

EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

**ACCELERATOR TARGETS DESIGNED
FOR THE
PRODUCTION OF NEUTRONS**



SYMPOSIUM
Liège, Belgium, September 18-19, 1967



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EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

PROCEEDINGS OF THE 3rd CONFERENCE ON
ACCELERATOR TARGETS DESIGNED FOR THE
PRODUCTION OF NEUTRONS

LIEGE (BELGIUM), SEPTEMBER 18-19, 1967

Edited by H.G. EBERT
Directorate General for Research and Training
Biology Division

*afab division.
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O P E N I N G A D D R E S S

L.J.F. WINAND
UNIVERSITE DE LIEGE

Mesdames, Messieurs,

J'ai l'agréable devoir de vous souhaiter la bienvenue à Liège où vous êtes réunis à l'occasion des colloques organisés par l'Euratom et l'Université de Liège et qui ont comme sujets de travail l'étude des cibles pour accélérateurs destinées à la production de neutrons d'une part et d'autre part les aspects pratiques de l'analyse par activation au moyen de particules chargées.

Vous avez répondu en très grand nombre à l'appel des organisateurs puisque 100 participants sont inscrits à la première partie du Colloque et 90 à la seconde, quinze pays sont représentés et je désire saluer spécialement ceux d'entre vous qui ont consenti à faire un très long voyage pour se joindre à nous.

Ce n'est certes pas pour les spécialistes que vous êtes qu'il faut souligner l'intérêt que présentent les deux sujets du colloque, mais il me plaît assez de rappeler que c'est en ce même Palais des Congrès, qui s'est tenu, pendant février 1964 le premier colloque consacré aux cibles pour accélérateurs, non pas dans cette grande salle, mais beaucoup plus modestement dans une salle de commission : nous étions 30 personnes qui avons très simplement tenté de faire le point des problèmes soulevés par la fabrication et par l'utilisation des cibles. Je suis très heureux de voir dans l'assistance la plupart

des participants à cette première réunion.

C'est à Grenoble que se tint en juin 1965 la seconde réunion de l'espèce et les organisateurs avaient cette fois jugé opportun de jumeler des discussions sur l'analyse par activation au moyen de particules chargées. Nous avons conservé de l'accueil de nos hôtes français un souvenir exceptionnel. Cette conférence avait rassemblé une centaine de participants et les exposés furent de très haut intérêt scientifique.

Sans nul doute le présent colloque rencontrera le même succès.

Avant d'ouvrir les séries d'exposés je désire remercier en votre nom l'Euratom et la Commission Administrative du Patrimoine de l'Université de Liège qui subventionnent le colloque.

L'organisation matérielle a été assurée en ce qui concerne l'Euratom, par le Dr. EBERT, du Groupe Radioisotopes de la Direction générale Recherches et Enseignement, à qui M. GODAR a passé le relai, en ce qui concerne l'Université de Liège, par le professeur J. Govaerts et ses collaborateurs, auxquels j'ai eu le plaisir de me joindre. Le centre d'Etudes de Chimie Métallurgiques du C.N.R.S. français s'est associé au patronage de la seconde partie de la conférence, je prierai mon collègue, le professeur ALBERT de transmettre nos remerciements au C.N.R.S.

Le Dr. EBERT, au titre de secrétaire général, vous donnera dans un moment quelques indications d'ordre pratique, tant en ce qui concerne le déroulement des exposés qu'en ce qui concerne les excursions et les visites. Je désire cependant avant de lui donner la parole, remercier la Société Philips, qui invite les membres de la conférence à prendre part à un voyage en autocar à Eindhoven mercredi prochain.

L'Université organise à l'intention des congressistes et des personnes qui les accompagnent, une réception qui aura lieu à la fin de cette première journée, à 18 H., à la Salle des Professeurs, place du XX Août, j'espère vous y accueillir très nombreux.

Mesdames, Messieurs, ma charge de doyen de la Faculté des Sciences ne me permettra pas d'assister à la clôture des débats vendredi. Je vous prie de m'en excuser et de m'autoriser à remercier par avance les présidents des séances, les orateurs, les secrétaires, les interprètes, nos hôtes étrangers et belges et tous ceux qui contribueront à la bonne marche et au succès de ce colloque. Tous les voeux de l'Université vous accompagnent.

Je déclare ouvert le colloque international Euratom 1967.

The Life of Tritium Targets under Deuteron Bombardment

D.L.E. SMITH

United Kingdom Atomic Energy Authority, Aldermaston

ABSTRACT

The Life of Erbium and Titanium tritide targets used in a 10mA, 300 KV accelerator has been measured. Various target holders are described and the cost of neutrons from tritium targets is discussed.

1. Introduction

At AWRE a 300 Kv Positive Ion Accelerator, made by the High Voltage Engineering Corporation, is being operated as a high intensity 14 MeV neutron source.

The cost of targets is a large item in the running costs of the machine and as a result the performance of targets has been assessed as usage of the accelerator has allowed. In this paper are described the accelerator performance, the types of target holders we use, the life test results and an estimate of the cost of running various target systems.

2. Accelerator Performance

Table 1 shows the accelerator performance. At 13mA, 350Kv the available ion beam power is over 4 kilowatts. This power can be concentrated on to a spot 4 cms in diameter. Power dissipation has been the main factor considered in the design of target holders for this accelerator.

3. Design of a water-cooled target assembly

Figure 1 shows a typical water cooled target holder for static targets, it takes a 2.5 cm diameter target. The holder is made of two concentric copper tubes and the target is soldered into the top of the inner tube. The water flow is shown by the arrows, it is guided across the back of the target by inserts marked 'A' in the top view. The inserts marked 'B' stop the water taking a short cut, the outlet is not shown.

With a water velocity of 5 metres a second across the back of the target at an inlet pressure of 5 kilograms per cm² the target will dissipate 1.5 kilowatts of incident ion beam. A 5 cm diameter version of this target which withstands 4 kilowatts of ion beam is also in use.

4. Rotary Targets

Figure 2 shows the rotor of a small rotary target holder which is available commercially. The target is a copper annulus 1.2 cm wide and

H.V.E.O. 300 KV ACCELERATOR PERFORMANCE

ION ENERGY	0 to 350 KeV
IONS.	D_1^+ 100% or D_2^+ 100%
Maximum DC Currents	D_1^+ 7mA D_2^+ 13mA
Maximum Pulsed Currents Pulses $\frac{1}{4}$ to 5 μ s at 100 to 2500 pps	D_1^+ 25mA D_2^+ 9mA

TABLE 1.

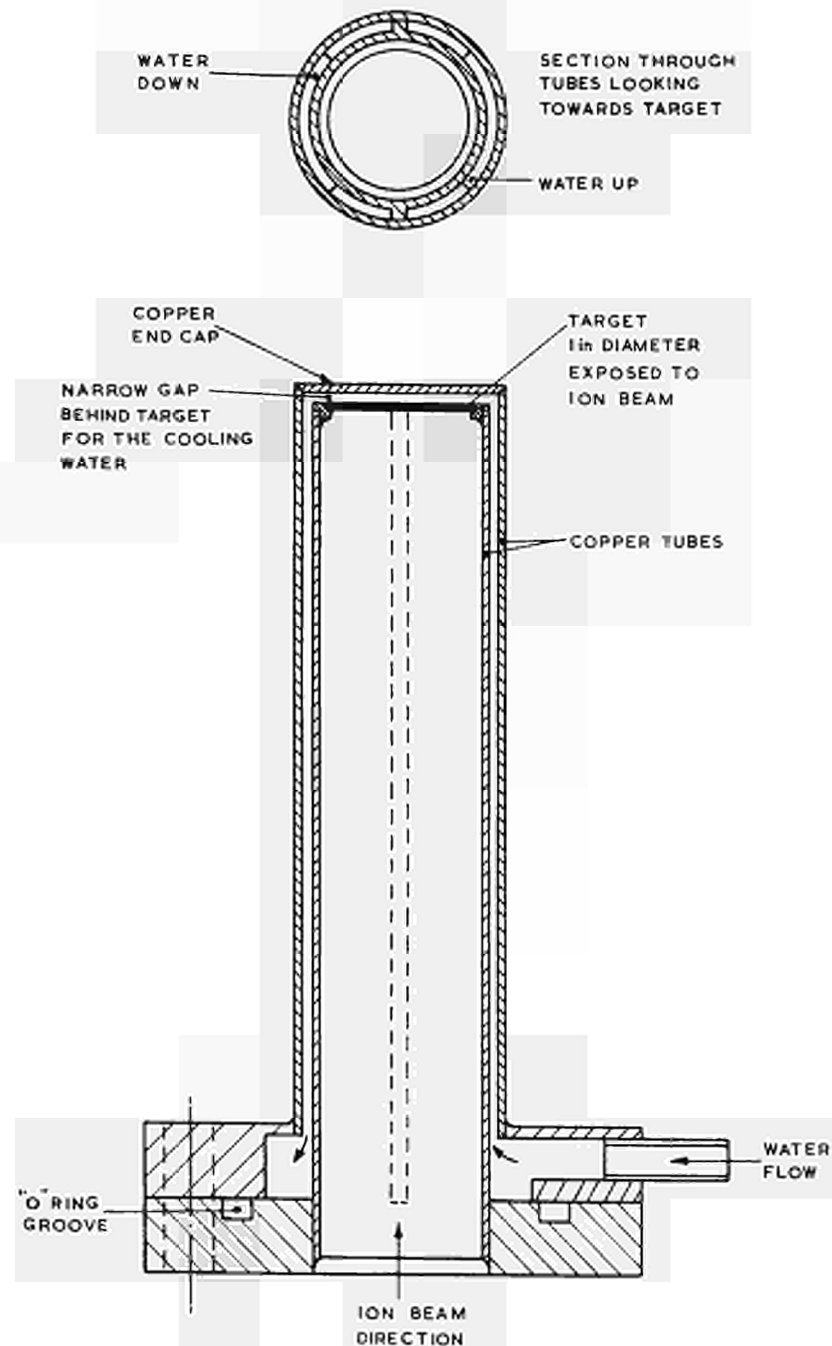


FIGURE 1. 'ONE INCH' TARGET ASSEMBLY SECTION

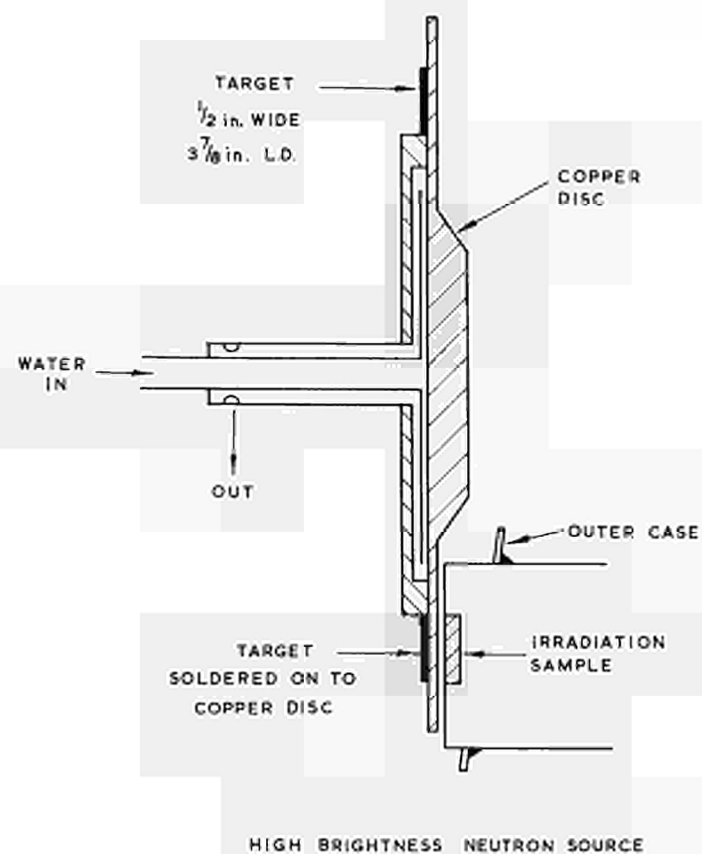


FIGURE 2. ROTOR OF 'MULTIVOLT' ROTATING TARGET

11 cm diameter with Titanium Tritide on one face. It is soldered on to the copper disc. As supplied by the manufacturer it will withstand 150 watts of ion beam without damage. We have increased its rate of rotation from 10 to 40 revolutions a minute and it operates satisfactorily at 600 watts. This power corresponds to a 14 MeV neutron flux of about 5×10^{10} n/cm²/sec in the sample position shown.

Figure 3 shows part of a rotating target designed by Dr.E.M. Gunnersen (AWRE) to withstand 3 kilowatts of ion beam. It was intended to give a source strength of 10^{12} neutrons a second for periods of up to 40 hours. The target consists of an annulus made up of 8 Molybdenum segments each five cms wide and 1 mm thick on a 40 cm diameter. There is a water filled cavity behind each segment. The disc is rotated once a second.

It was first tested with Deuterium targets to make sure it would withstand 3 kilowatts of ion beam without deteriorating. Erbium Tritide targets were then mounted on the rotor and it was operated until the neutron output fell to one half the initial output.

5. Results of Operating Tests

5.1. Figure 4 shows a typical life test result.

The neutron yield falls at a steady rate from its initial value and the point at which the neutron yield reaches one half the initial yield is a convenient reference. That is point 'A' on the diagram.

$$\text{Target life} = \frac{\text{milliamps of ion current} \times \text{hours to half yield}}{\text{target area in square centimetres}}$$

These are convenient dimensions which relate to accelerator running time and the definition is applicable to any target.

The example shown had a life of 2.5 mA hours per square centimetre. The effect of Deuterium build up is also shown, (right hand scale). At the point where the curves cross the 3 MeV neutron output is 1% of the 14 MeV neutron output. The D(T) and D(D) neutron outputs were separated by using a semi-conductor counter which measured the alpha and proton emissions from the target. The counter had a thin foil in front of it to reduce the alpha energy and thus separate it from the proton.

The majority of life tests have to be made at high neutron outputs which make it difficult to use alpha counters. Fission chambers monitoring

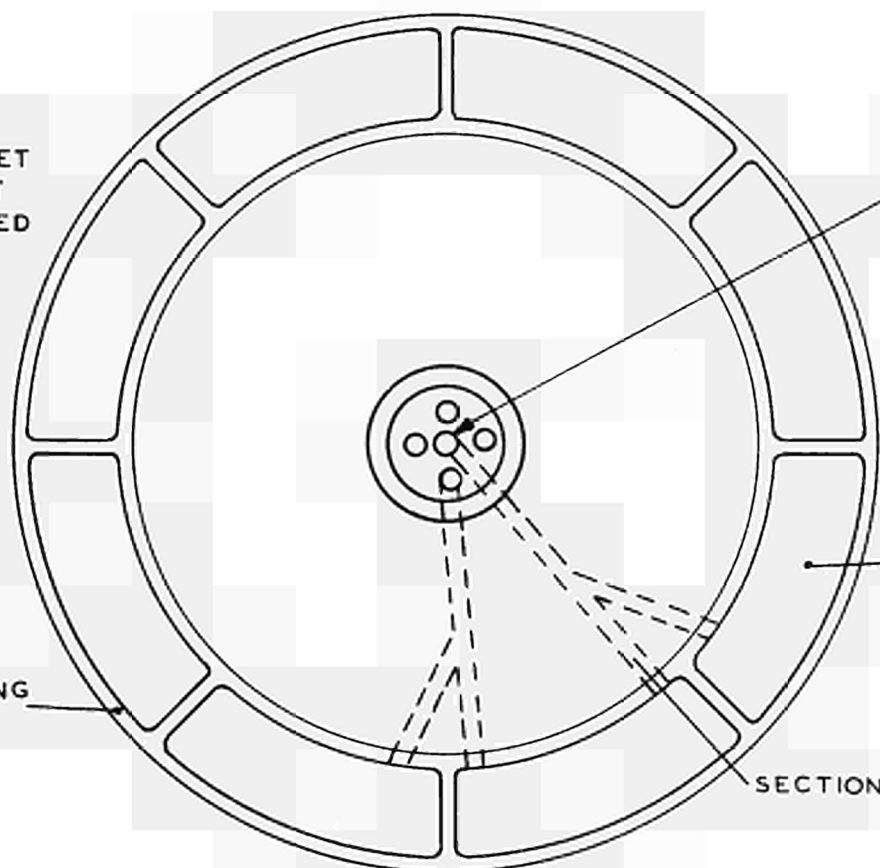
ROTOR PLAN
SHOWING ONE INLET
AND ONE OUTLET
FOR WATER, DOTTED

CLAMPING
RING

WATER
INLET

TARGET
SEGMENT

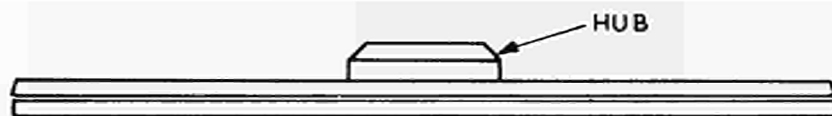
SECTION A



ROTOR
SIDE
ELEVATION

HUB

ROTOR
BODY



ION BEAM
DIRECTION

CLAMPING RING
SHOWING
SCREW HOLE

OUTER
CASE

ROTOR
BODY

WATER
FLOW

2 in

TARGET

WATER CHEST

IRRADIATION
RECESS

SECTION 'A'
SHOWING OUTER
CASE AT BEAM
INLET POSITION

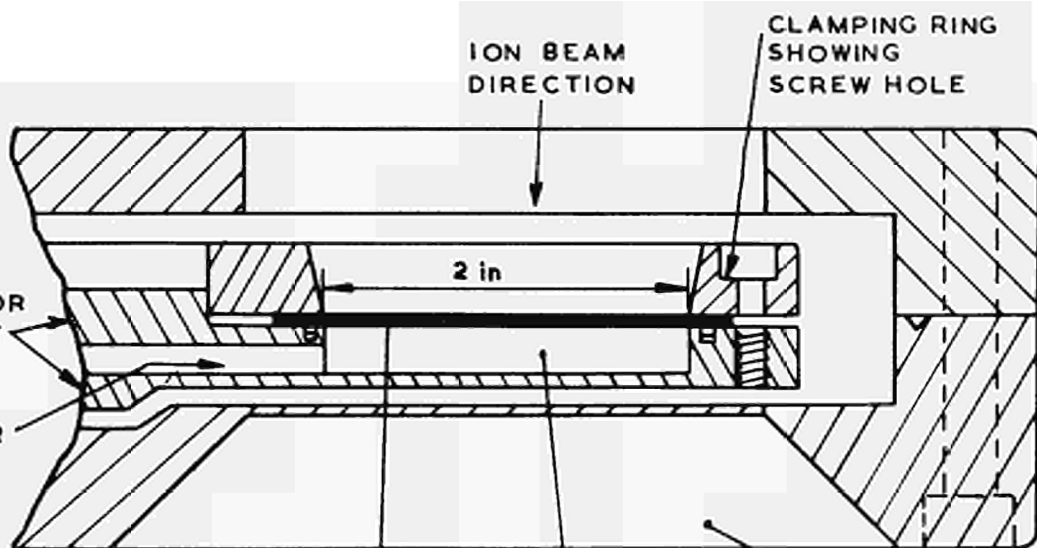


FIGURE 3. ROTATING TARGET ASSEMBLY

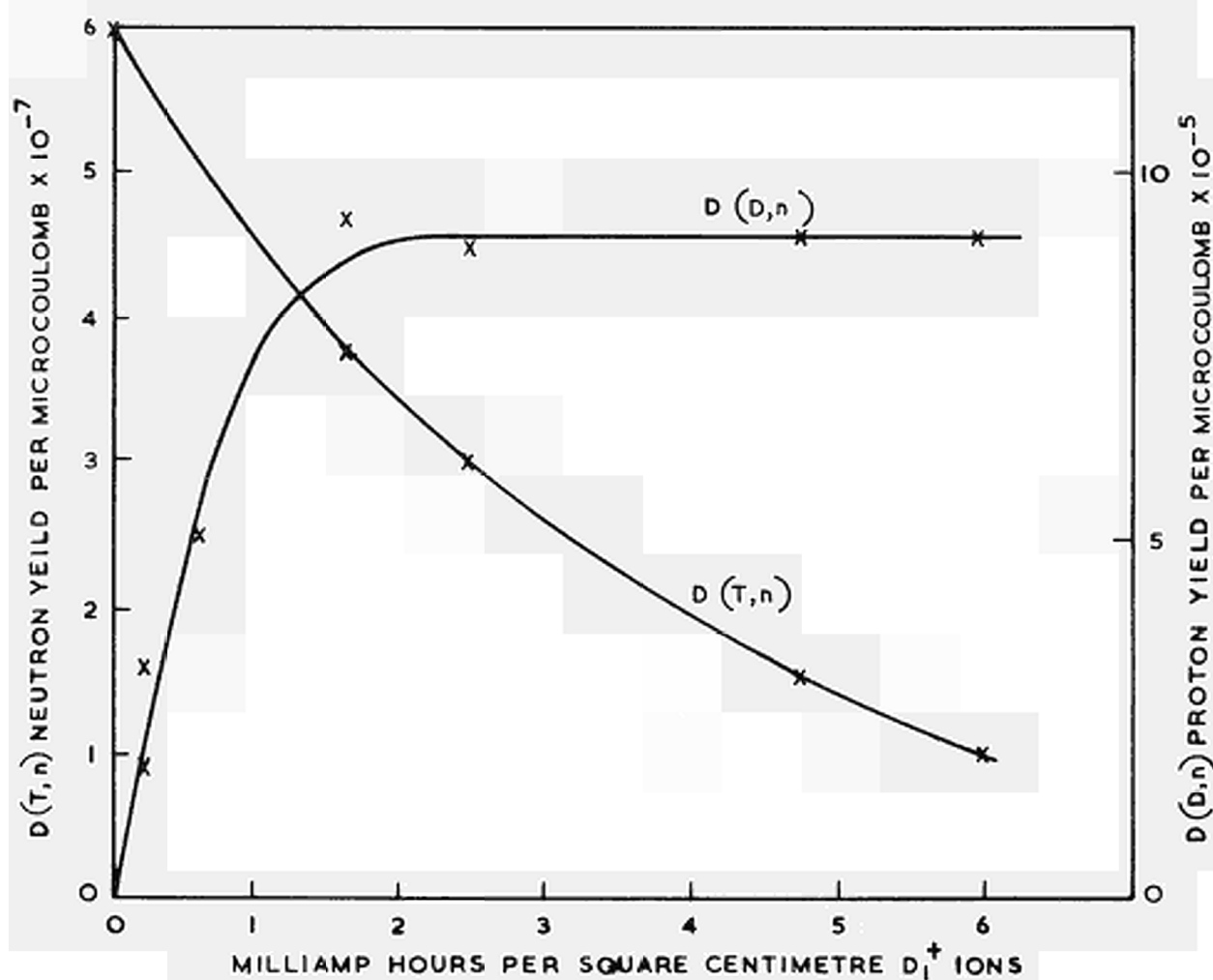


FIGURE 4. NEUTRON YIELD VERSUS INTEGRATED CURRENT FOR A CENTRAL 0.9 cm DISC ON A 1.1 cm DIAMETER TARGET AREA EXPOSED 0.636 cm²

the neutrons directly were used in these cases.

5.2. The complete test results are shown in Table 2. The first four results refer to targets made at the Radiochemical Centre, Amersham, England.

Result 1: This refers to tests on the targets normally used and is an average of about 20 tests over a wide range of ion currents. About one in ten targets has to be rejected because of low neutron yield and these are not included in this result.

Result 2: The object of this test was to see if vacuum conditions and cleanliness affected the target life. We used a target holder in which the target rested on 'O' ring seals, this avoided getting solder flux on the target surface. After loading the target on to the accelerator we waited until the pressure reached 1×10^{-6} torr and kept the pressure below 2×10^{-6} torr during the test. The power incident on the target was 340 watts per square centimetre and its temperature rose to about 100°C during the test. Afterwards the target surface was found to be pitted and rough.

Result 3: This is the result of testing the small rotary target. The target is designed to give a high neutron flux over a very small volume, and a life of 100 mA hours. We have not used it enough to obtain an accurate life figure but results so far indicate that it may not differ from the figure in Result I.

Result 4: This result shows that these Erbium Tritide targets compared poorly with corresponding Titanium Tritide targets.

Result 5: This shows the result of the life test on the large rotary target. The initial output was close to 10^{12} neutrons a second but this was not maintained, falling to half in thirty hours on the accelerator.

6. Comparative Cost of Targets

On the basis of the above results an estimate has been made of the target running costs based on a 2.7 mA hour life. The two applications considered

TRITIUM TARGET TESTS

TARGET TYPE	Typical initial neutron yield per microcoulomb at an ion energy $E_{D_2}^+ = 300 \text{ KeV}$	Half Life mA hours per cm^2	Range of Target Currents used in tests. Milliamps.
Titanium on Copper Ti. thickness 2.5 mg/cm 1.0, 2.5 and 5 cm dia.	1.5×10^8	2.7 ± 0.8	0.1 to 13
Titanium on Copper Idealised treatment (one only) Ti. 2.5 mg/cm^2 centre portion of 2.5 cm dia. target.	1.9×10^8	7.7	0.8
Titanium on Copper Rotating Assembly. Ti. 2.5 mg/cm^2 Annulus 5.5 cm. rad. 1.2 cm wide	1.5×10^8	Greater than 2.0	0.1 to 2.0
Erbium on Chromium Plated Copper. 2 samples. Er. 7 mg/cm^2 2.5 cm dia.	0.8×10^8	0.1	1 to 2
Erbium on Molybdenum Rotating Assembly Er. 7 mg/cm^2 Annulus 20cm. radius. 5 cm wide.	1.01×10^8	0.3	7 to 9

TARGET ATOMIC RATIOS	TRITIUM TO TITANIUM	1.5 to 1
	TRITIUM TO ERBIUM	1.7 to 1

TABLE 2.

were, first, the target cost per neutron produced and second, the target cost per unit neutron flux through a small sample. The costs are listed in Table 3.

The first two target systems were designed for applications where target cost is not important. The last three target systems were designed for high neutron output. It is clear that in this case the choice of the most economical target depends strongly on the particular application.

Further details of this work can be found in AWRE report O - 52 - 67 which has recently been published.

Accelerator targets designed for the production of Neutrons

Additional Notes by D.L.S. SMITH

Some target users may not be aware of the limitations on the efficiency of the different target cooling systems shown at this conference.

There are two basic methods of removing heat from a target with water these are:-

1. Transfer of the heat directly from the back of the target support into the water.
2. Conduction of the heat through the target backing with water remote from the target.

Method 1

We assume that the target support is thin enough (dimension t in Figure 5) that the heat loss to the edges can be ignored. Also if the maximum permissible temperature rise is 100°C radiation losses are negligible. The only way of removing heat from the target is then by movement of the water across the back of the target support. Several speakers have said, power dissipation in excess of 500 w/cm^2 can be obtained provided the water velocity is sufficiently high to give turbulent flow and prevent bubble formation. I have found that the following empirical method of determining the required flow in a target holder can be used to guarantee adequate cooling for dissipations up to 400 watts/cm^2 . The temperature rise of the water must not exceed 5°C and the velocity must exceed 5 metres/second. Knowing the total heat input the volume flow required can be calculated.

If the depth of water behind the target is greater than one or two millimetres it can be difficult to supply the volume of water required to

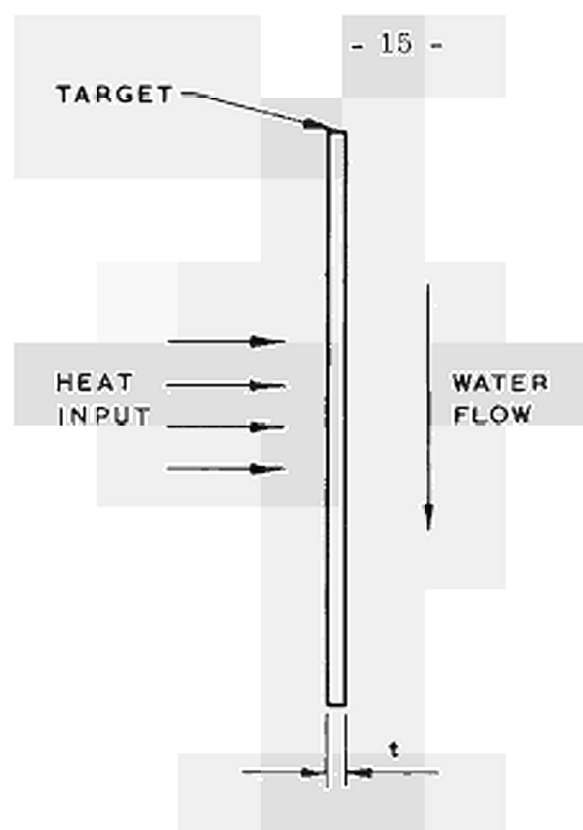


FIGURE 5.

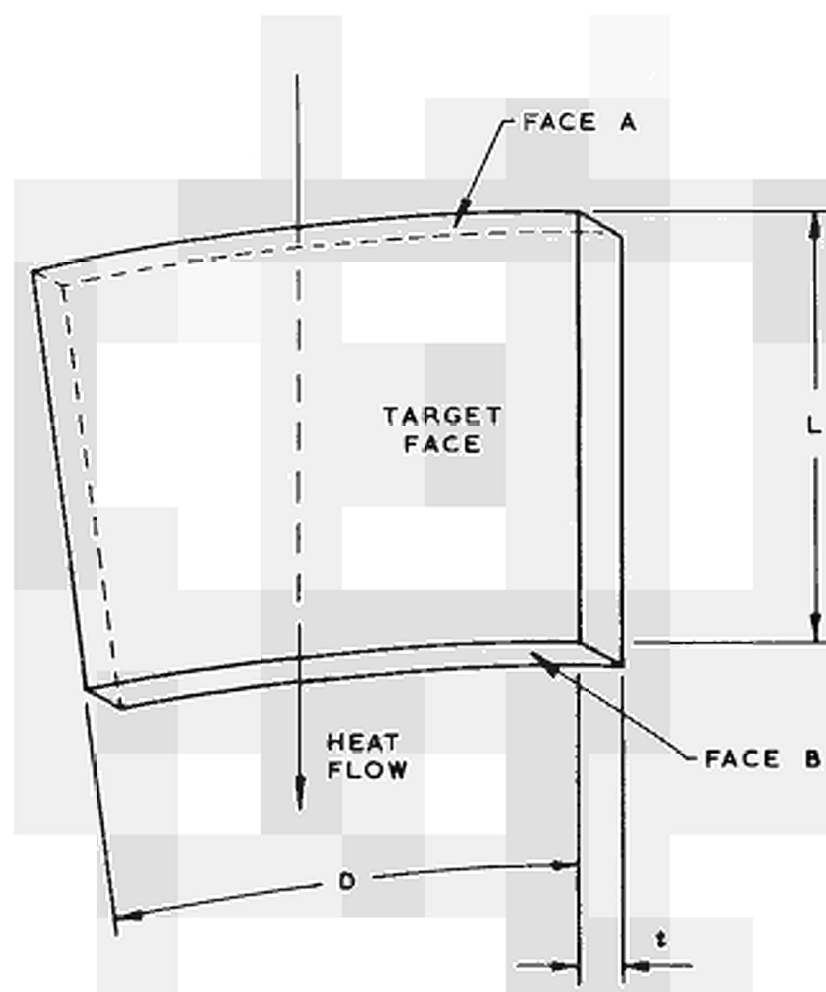


FIGURE 6.

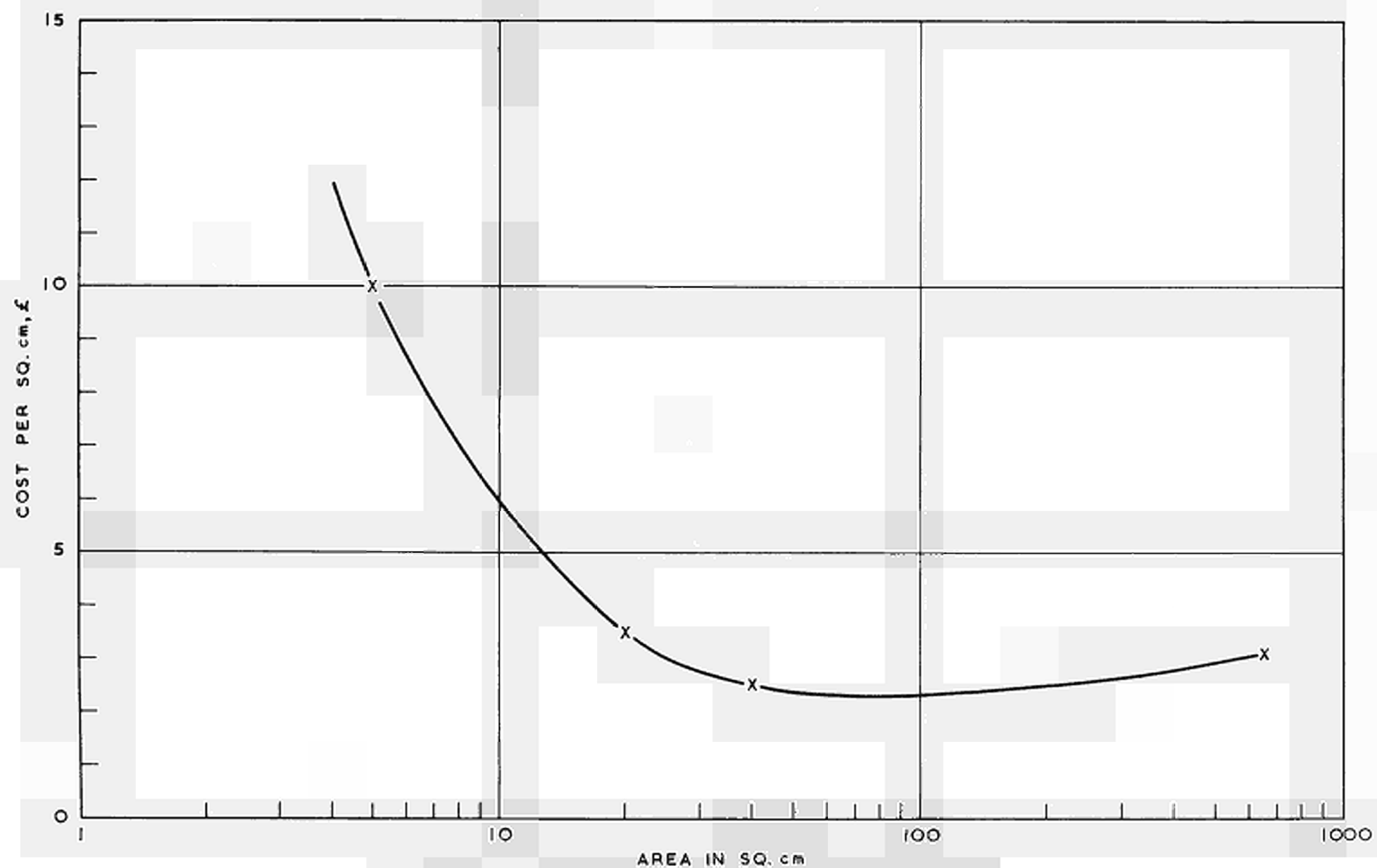


FIGURE 7. COST OF TARGETS USED AT A.W.R.E.

TRITIUM TARGET RUNNING COSTS

TARGET TYPE AND SIZE	TARGET COST	TARGET COST PER SQU. CM	COST PER 10^{14} SOURCE NEUTRONS	MINIMUM COST PER 10^{14} n/cm ² THROUGH SMALL SAMPLE
	£	£	£	£
FIXED 1 cm.	35	35	4.9	25
FIXED 2.5 cm.	50	10	1	17
FIXED 5.0 cm.	70	3.5	0.3	20
SMALL ROTARY 11 cm.	100	2.4	0.37	1.85
LARGE ROTARY 40 cm.	2000	3.2	0.55	27.5
ROTATING TARGET MINIMUM COST AT LIFE OF 2.7 mA Hours/cm ²			0.25	

These costs include depreciation of the target holder.

TABLE 3.

obtain the velocity. If the depth of water is too small there may be insufficient water to keep the temperature rise below 5°C unless excessive pressure is used. Water pressures of 7 Kg/cm² are required to obtain these conditions in the target shown in Figure 1. Some of the target holders shown at this conference would not withstand such pressure.

With a thin target support there is no appreciable gain in permissible heat dissipation by using a rotary system. If the support is thickened then heat is stored in the support which can be transferred to the water while the ion beam is not on that part of the target behind it and a gain in power dissipation is obtained.

The method of calculating target temperature in this case is given in my AWRE report quoted at the end of my paper.

Method 2

In Figure 6 is shown a sector of a rotary target of thickness t where the heat is transferred in the direction of the arrow from A to B. If it is assumed that all the heat is incident at the upper face A, then the maximum power dissipation can be calculated from

$$P = \frac{T \times L \times 4.18}{C \ t \ D}$$

Where P = Watts
T = temperature difference
between A and B
C = Heat conductivity

This serves as a pessimistic estimate of the power dissipation of the edgecooled rotary target where the heat (ion beam) is applied at the target face shown. If the face B fits round a heat exchange system then a upper limit to the power dissipation for a given temperature rise at face A can be found by assuming an infinite rotating speed which would give uniform heating of the target. If D is much greater than L, which is true for most currently available rotating targets, then the ratio of the maximum heat dissipations in the rotating and stationary conditions is a little less than D/L. This is because the heat flow is no longer radial when the target is stationary or revolving slowly. (It is assumed that the ion beam diameter equals L).

Target cost

The cost of the tritium targets we use does not vary linearly with area, Figure 7 shows the cost as a function of area. It is plain that large area targets can be more economic than the small area ones in terms of the cost of neutrons produced provided full use can be made of the target holder.

DISCUSSION

Mr. JIGGINS

Dr. SMITH mentioned that the output of his machine, under continuous running conditions, was found to be reduced by about 90% when it had not been used for a long period. We also have had this experience. I would like to ask what he did to regain the full output, **was it** simply a question of retuning the ion source ?

Mr. SMITH

After a long period in which the accelerator was not operated near its maximum output (1 year), it was found that the ion beam current available had decreased by a factor of 3 to 5. In order to regain the ion current output it was necessary to run the accelerator for two or three days at its maximum output. During this time the ion source controls were varied in order to keep the ion current as high as possible. At the end of two days the output was back to near its original maximum.

UNE METHODE DE DETERMINATION DE LA DISTRIBUTION
SPATIALE DU TRITIUM DANS LES CIBLES

A SUPPORT MINCE

E. FORT, J.L. HUET

CEN CADARACHE

A METHOD OF DETERMINING THE SPATIAL DISTRIBUTION
OF TRITIUM IN THIN-BACKED TARGETS

ABSTRACT

The method consists essentially of determining the energy spectrum of charged He^3 particles associated with neutrons in the T(p,n)He^3 reaction.

The He^3 particles produced at a fixed angle, ten degrees towards the front, by a proton beam of constant energy, are separated by an electromagnetic analyzer from the particles coming from parasite reactions.

They are detected by a surface-barrier gold-silicon detector.

By knowing the range-energy curves for the He^3 particles, deduced from the range-energy curves for the protons in the target material, it is possible to arrive at the tritium distribution in the target from the energy spectrum of the He^3 particles.

The accuracy of the measurements depends primarily upon that of the range-energy curves for the He^3 particles and upon the resolution of the analyzer.

INTRODUCTION

La connaissance de la distribution du tritium dans les cibles Ti-T à support mince destinées à la production de neutrons a déjà fait l'objet de plusieurs expériences.

GUNNERSEN et JAMES (1) ont montré que dans le cas d'une cible Ti-T il y a toujours en surface une couche non tritiée et une diminution assez rapide de la densité de tritium vers l'autre extrémité du dépôt de titane.

Des résultats similaires ont été obtenus par MACKLIN et GIBBONS (2), FIELDHOUSE et al. (3).

Pour nous, utilisateurs de cibles tritiées, le problème de la répartition du tritium revêt une grande importance.

La nécessité de connaître la forme de cette distribution nous est apparue de façon précise lors de l'étalonnage d'un scintillateur au verre de Li (NE 905), pour des neutrons d'énergie inférieure à 100 KeV. Cet étalonnage est effectué par la méthode de la particule associée. L'efficacité absolue du scintillateur est le rapport du nombre de coïncidences $n\text{-}^3\text{He}$ au nombre de particules ^3He . (4).

DESCRIPTION DES CIBLES ET DE L'EXPERIENCE

Les trois cibles étudiées sont des cibles Ti-T d'environ $200 \text{ } \mu\text{g}/\text{cm}^2$ d'épaisseur à support mince de cuivre ($400 \text{ } \mu\text{g}/\text{cm}^2$)

Leur fabrication est réalisée par le Bureau Central des Mesures Nucléaires à Mol en Belgique, tandis que l'imprégnation en tritium est faite au département des Radioéléments de Saclay.

Le rapport T-Ti est de l'ordre de 1/1 dans les cas les plus favorables. Toutefois ce rapport n'a pas été donné avec précision par le fabricant.

Leurs caractéristiques sont les suivantes :

N° cible	Epaisseur Ti $\mu\text{g}/\text{cm}^2$	Support Cu $\mu\text{g}/\text{cm}^2$	Remarques
1	220,5	406,6	P t
2	198,6	401	
3	177,5	423	P t

Deux de ces cibles (n°s 1 et 3) ont un mince dépôt (entre 0,5 et 1,2 $\mu\text{g}/\text{cm}^2$) de platine entre le support de Cu et la couche de Ti. Le rôle de ce dépôt est d'empêcher la diffusion du tritium dans le cuivre lors de l'imprégnation et de l'utilisation de la cible. (5).

Les particules ^3He , produites vers l'avant à 10 degrés par rapport au faisceau incident de protons, sont collimatées avant de pénétrer dans l'ensemble analyseur.

Celui-ci se compose d'un analyseur électrostatique donnant une déflexion des particules chargées, proportionnelle au rapport z/E de la charge et de l'énergie de la particule, et d'un analyseur magnétique donnant une déflexion inversement proportionnelle à la quantité de mouvement et proportionnelle à la charge.

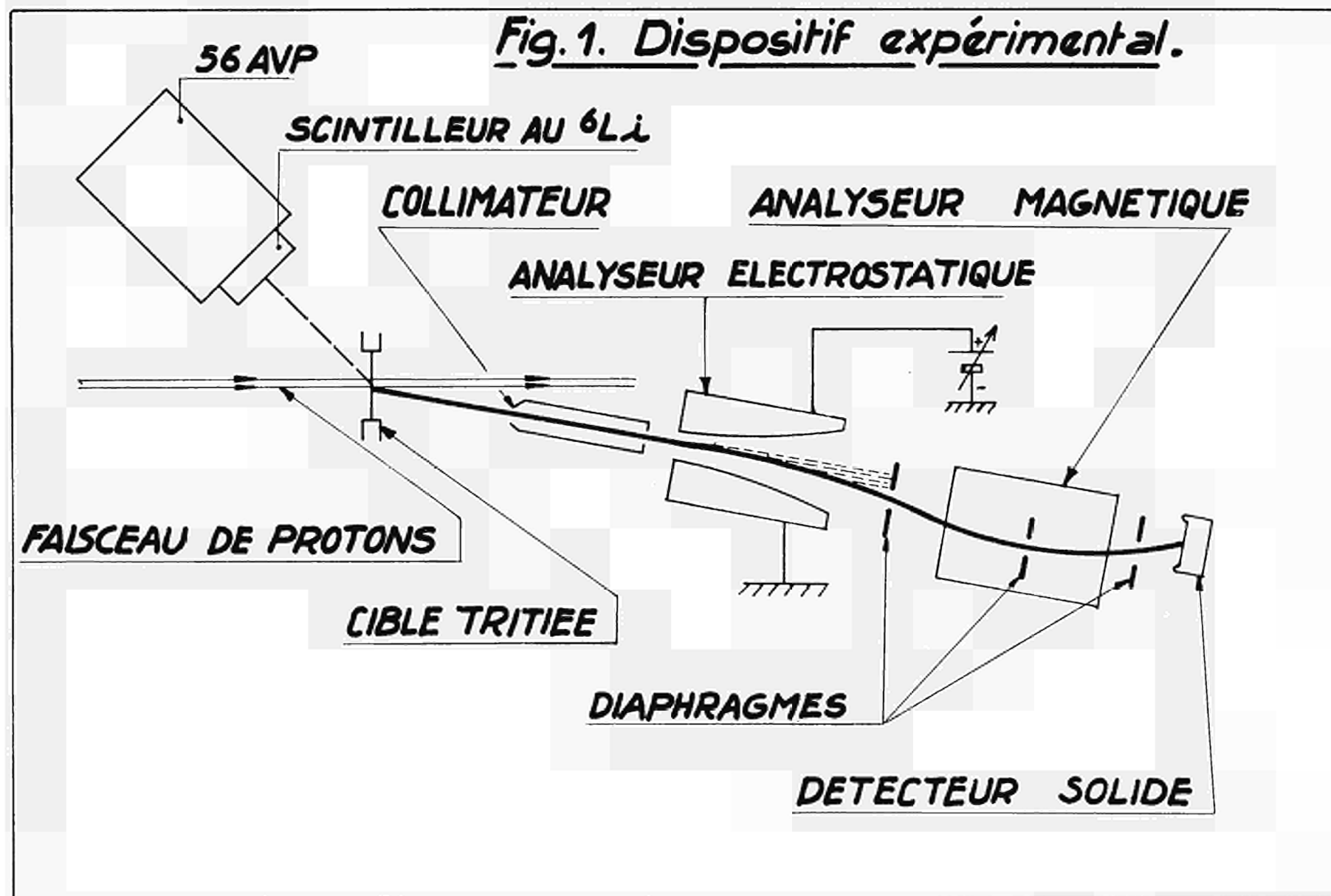
L'ensemble de ces deux analyseurs fonctionne donc en spectrographe de masse et on vérifie que le taux de particules parasites résiduelles (protons inélastiques, tritons de recul etc....), est inférieur à 3 %. De toutes façons ces particules parasites transmises ne constituent pas une gêne car elles sont facilement identifiées sur les spectres.

Les particules ^3He sont ensuite détectées par un détecteur or-silicium à barrière de surface (type ORTEC/5 - 532 A) ayant une fenêtre insensible d'or de 93.5 $\mu\text{g}/\text{cm}^2$ d'épaisseur. Le schéma de l'expérience est donné sur la figure 1.

On fait varier simultanément les paramètres de déflexion, c'est-à-dire V , la tension appliquée aux plaques du séparateur électrostatique, et i le courant d'induction dans le séparateur magnétique, de façon que la variation relative de celui-ci soit égale à la racine carrée de la variation relative de la tension V (6).

De cette façon on sélectionne successivement les particules ^3He produites dans la cible à différentes profondeurs.

Pour étalonner la chaîne électronique linéaire nous nous sommes servis d'une source de Plutonium-Uranium délivrant deux raies de particules alpha d'énergie 5,147 MeV et 4,816 MeV.



EXPLOITATION ET DISCUSSION DES RESULTATS EXPERIMENTAUX :

Les différents spectres énergétiques obtenus sur un analyseur à 400 canaux, apparaissent relativement larges, leur largeur à mi hauteur étant d'environ 90 KeV.

Comme les particules ${}^3\text{He}$ ont une énergie maximum de 992 keV lors de leur production, la résolution en énergie de l'ensemble chaîne électronique et analyseur est de l'ordre de 10 %.

Cette résolution doit être attribuée essentiellement à l'ensemble collimateur d'entrée et analyseur.

On suppose que la différence ΔE entre l'énergie des ${}^3\text{He}$ lors de leur création, et leur énergie mesurée, provient entièrement de leur perte d'énergie dans l'épaisseur de cible X_r qu'ils traversent et dans la zone insensible du détecteur

$$\Delta E = E_{{}^3\text{He th}} - E_{{}^3\text{He mes.}} = \Delta E_{\text{dit}} + \left(\int_0^{X_r} \frac{dE}{dx} dx \right)_{\text{cible}}$$

Les pertes d'énergie différentielles dE/dx relatives aux particules ${}^3\text{He}$ sont dérivées des pertes d'énergie dE/dx relatives aux protons par la relation

$$\epsilon_{{}^3\text{He}}(E) = 4 \epsilon_p(E/3)$$

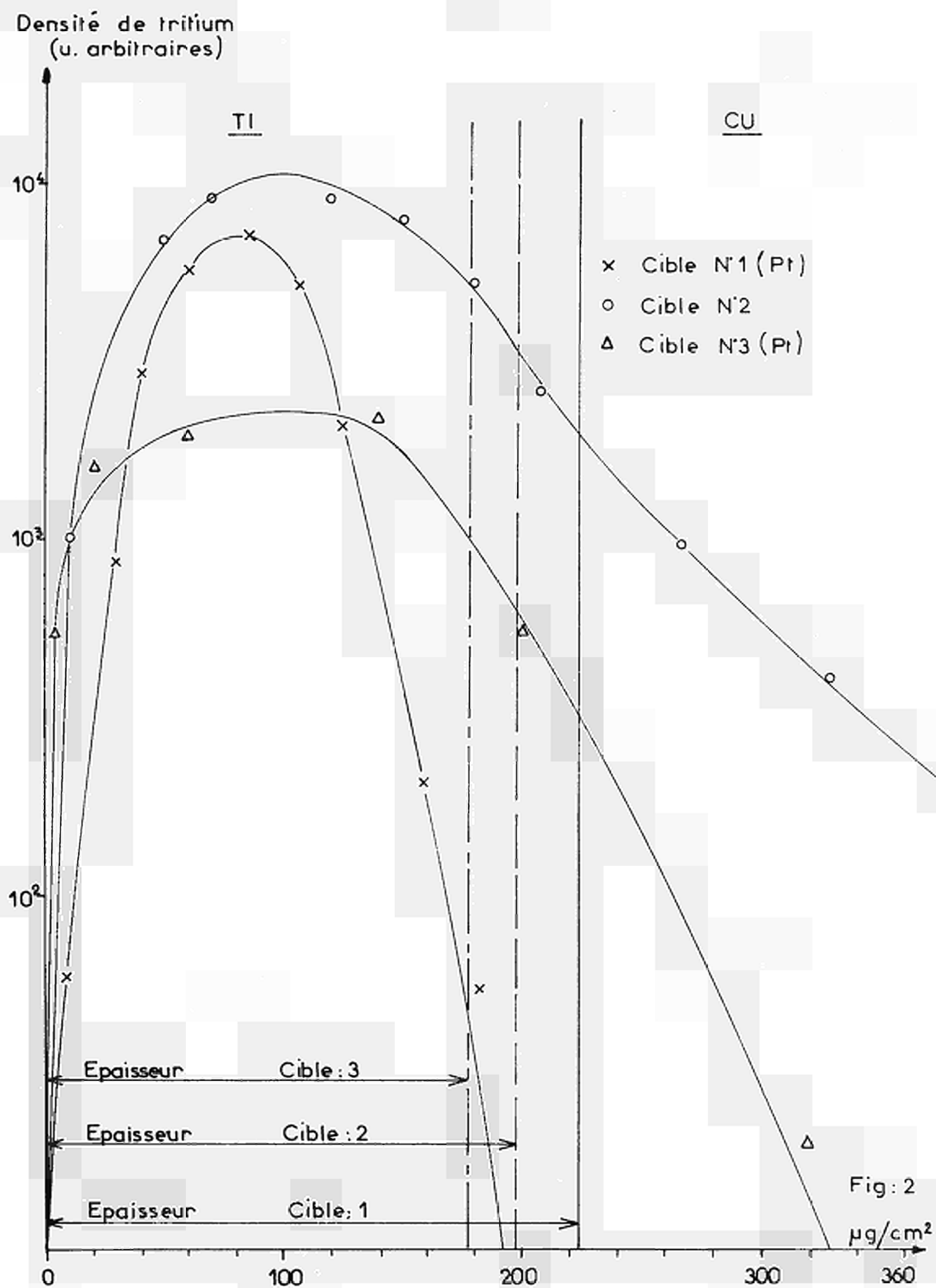
Les sections efficaces de ralentissement ϵ_p des protons dans le titane deutéré sont celles publiées par W. WHALING (7,8)

La procédure suivie pour calculer la perte d'énergie des particules ${}^3\text{He}$, d'énergie relativement faible, est entachée d'une erreur assez importante, pouvant aller jusqu'à ± 20 %.

Nous ne disposons malheureusement pas de courbes de pertes d'énergie pour des particules ${}^3\text{He}$ d'aussi faible énergie.

La courbe donnant le nombre de particules ${}^3\text{He}$ en fonction de l'épaisseur de la cible est aussi celle qui donne la distribution du tritium, puisque la section efficace de la réaction $T(p,n){}^3\text{He}$ est pratiquement constante dans toute son épaisseur.

Les résultats obtenus sont portés sur la figure 2 en échelles semi-logarithmiques.



En premier lieu nous pouvons constater que les distributions de tritium diffèrent considérablement d'une cible à l'autre.

Mais la forme de ces distributions présente toujours un front raide vers la surface avec une couche non tritiée inévaluable dans nos mesures, et une diminution plus lente de la densité de tritium vers l'autre extrémité.

La présence du dépôt interstitiel de platine entre le titane et le support de cuivre pour les cibles n° 1 et 3 n'empêche apparemment pas le tritium de diffuser dans le cuivre.

Sur la figure 3 sont portées deux distributions de tritium pour une même cible mesurées avant et après une irradiation prolongée. Les courbes ont été tracées en supposant que le bombardement prolongé n'avait pas eu d'effet sur l'épaisseur de titane. On constate nettement, notamment par le tableau 3, la dégradation de la densité de tritium dans la cible après une longue utilisation.

Ce phénomène est dû essentiellement à la diffusion du tritium en dehors de la cible ; sa destruction sous l'effet des réactions nucléaires étant négligeable.

CONCLUSION

En conclusion, nous pensons que cette méthode, bien qu'elle mette en oeuvre un appareillage assez complexe, nous permet d'avoir une idée assez réaliste de la distribution du tritium dans les cibles à support mince.

La principale incertitude provient du manque de précision des courbes de pertes d'énergie relatives aux particules ^3He .

Bien entendu les résultats que nous présentons sont trop fragmentaires et notre effort dans ce domaine doit être poursuivi, si possible avec des particules de plus grande énergie.

Nous croyons tout de même intéressant de souligner le fait que le tritium diffuse dans le support et surtout que les distributions obtenues diffèrent toutes les unes des autres.

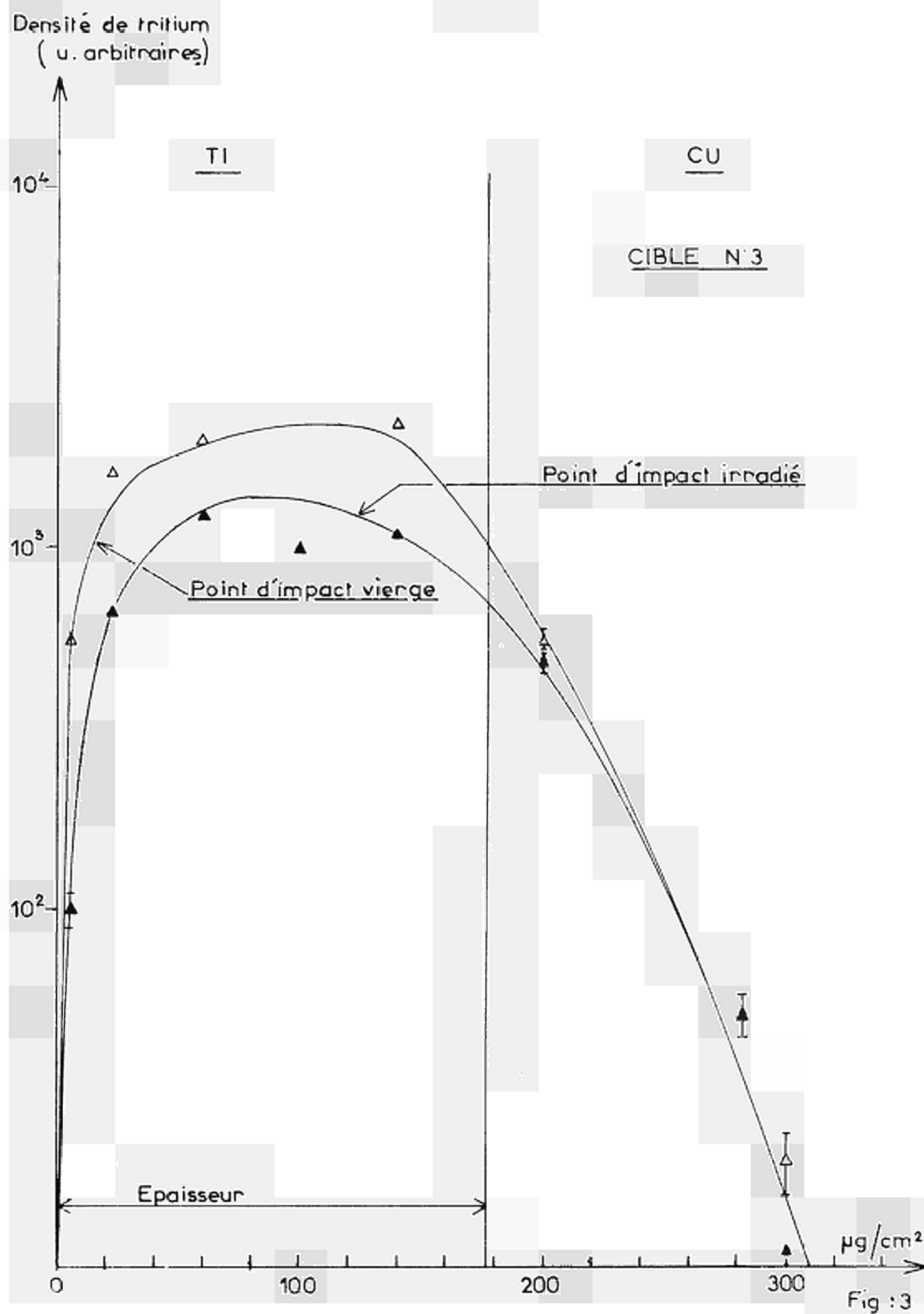


TABLEAU III

e / μ g/cm ²	R
0	8
5	5
22	2,3
60	1,58
140	1,98
200	1,14

Rapport des concentrations en tritium
avant et après l'irradiation.

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DISCUSSION

Mr. QUAGLIA

Savez vous comment a été appréciée l'épaisseur de 5 $\mu\text{g}/\text{cm}^2$ de Pt entre le Ti et le Cu ?

Mr. FORT

Je suis dans l'impossibilité de vous répondre. J'avais exprimé le désir d'avoir des cibles de Ti/Cu avec une couche intermédiaire de Pt d'une épaisseur voisine de celle d'une couche moléculaire. N'ayant reçu aucune indication particulière, j'ai fait entière confiance au constructeur qui est le service de Mr. **DEBUS** du BCMN de Geel (Belgique) , auprès de qui vous trouverez sans doute toutes les précisions demandées.

LA FABRICATION DE CIBLES TRITIÉES à SACLAY

A. MANIN et D. CHOLET
CEN SACLAY

ABSTRACT

THE FABRICATION OF TRITIATED TARGETS AT SACLAY

Tritium targets are prepared at Saclay by a conventional method consisting of two well-separated stages, namely, the volatilization of titanium and the impregnation of the thin layer.

A description is then given of the main controls of our machines, their empirical nature being stressed. Controls are never transposed from one apparatus to another without correction.

Lastly, the maximum titanium thickness and tritium charge attainable are given. Brief reference is made to the main faults which may occur on the targets.

INTRODUCTION

L'utilisation principale des sources de tritium adsorbé sur Titane est, en France, la production de neutrons par des accélérateurs.

On fabrique, pour cet usage, des cibles dont l'épaisseur de titane est 0,300 à 1,00 mg/cm² (0,7 à 2 microns) et le contenu en tritium de 0,1 à 0,4 millilitres par centimètre carré.

Nous fabriquons aussi des sources de rayonnement β pour l'ionisation des détecteurs de chromatographie. Ces sources doivent avoir une épaisseur inférieure à 0,25 mg par cm². Le courant utilisable n'est pas prévisible. Il dépend de plusieurs paramètres qu'il est malaisé de fixer.

Nous fabriquons enfin des sources de rayons X pour l'excitation de la fluorescence. Ces sources sont réalisées en général avec du zirconium. Elles sont plus épaisses que les cibles neutrogènes.

MODE DE PREPARATION

Nous travaillons, à Saclay, en deux opérations principales, réalisées dans des laboratoires différents, suivant des procédés classiques et connus.

1°/ La volatilisation du titane pour obtenir une couche mince est faite dans un évaporateur assez grand.

2°/ L'imprégnation des cibles est faite dans des appareils ne contenant qu'une cible (figure 1).

VOLATILISATION

Les supports d'argent, de cuivre, d'acier inoxydable sont dégraissés, grattés au papier émeri ou sablés, nettoyés par étuvage sous vide (300° C 10⁻⁴ Torr) ; ensuite pesés et chargés dans l'évaporateur.

Le titane est chargé dans les creusets de tungstène sur la platine de l'appareil à évaporer (figure 2). Les creusets contiennent de 50 à 500 milligrammes de titane, suivant l'épaisseur désirée.

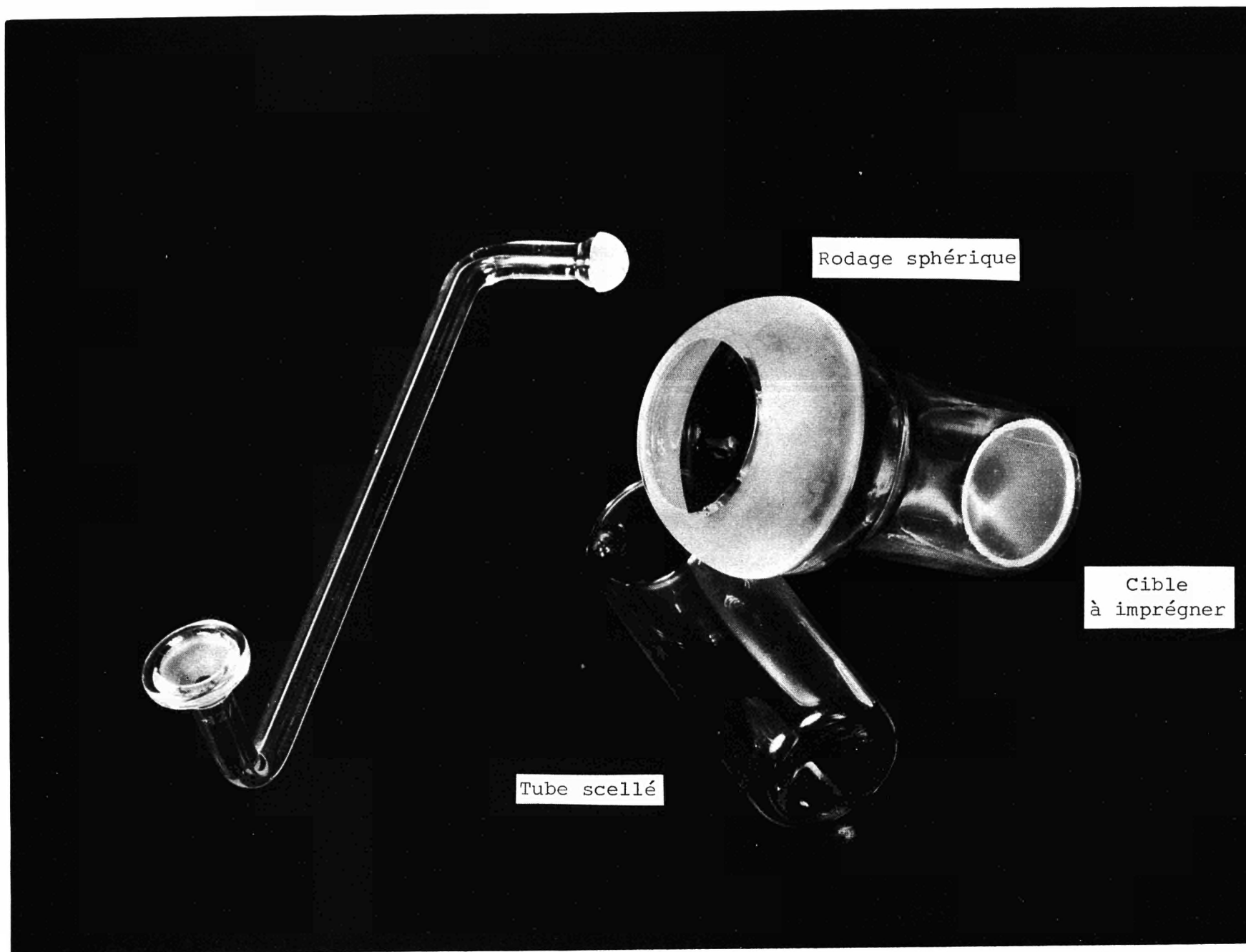


Figure 1 : récipient d'imprégnation

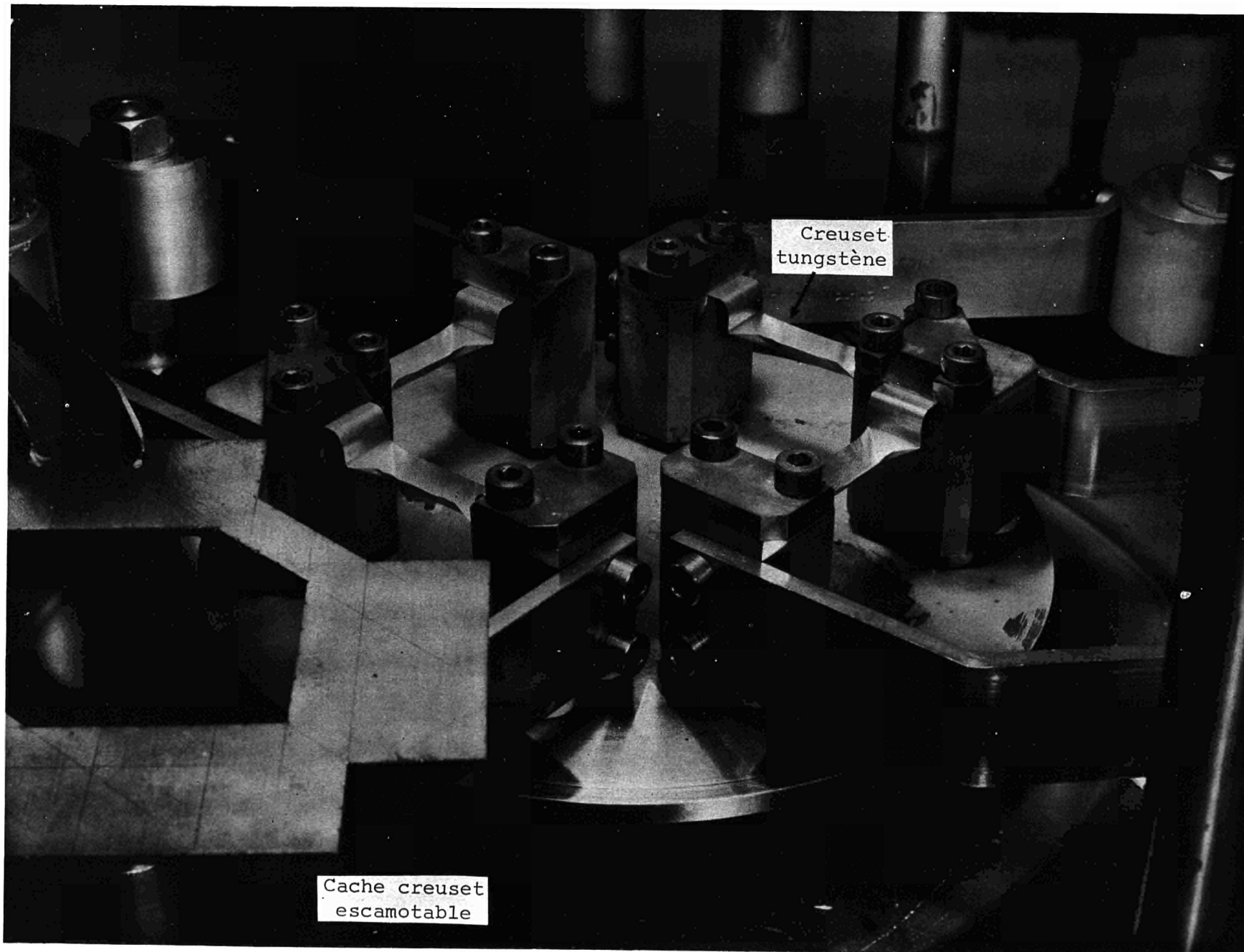


Figure 2 : platine d'évaporation à quatre creusets.

Le dégazage préliminaire déterminé empiriquement pour chaque appareillage est, sur l'appareil qui vous est présenté, de 3 heures à 250° C environ ; les creusets contenant le titane sont, eux, chauffés pendant ce temps à 800°/900° C environ.

La puissance de chauffe est déterminée empiriquement, de manière à avoir le minimum de dégazage au moment de la fusion du titane.

La volatilisation se fait en chauffant successivement les quatre creusets au point de fusion du titane. On augmente le chauffage au moyen d'un autotransformateur variable.

La figure 3 représente la variation de pression pendant la volatilisation des quatre creusets.

On laisse refroidir l'appareil toute la nuit ; au matin, les supports sont pesés et envoyés à l'imprégnation.

COMMENTAIRES

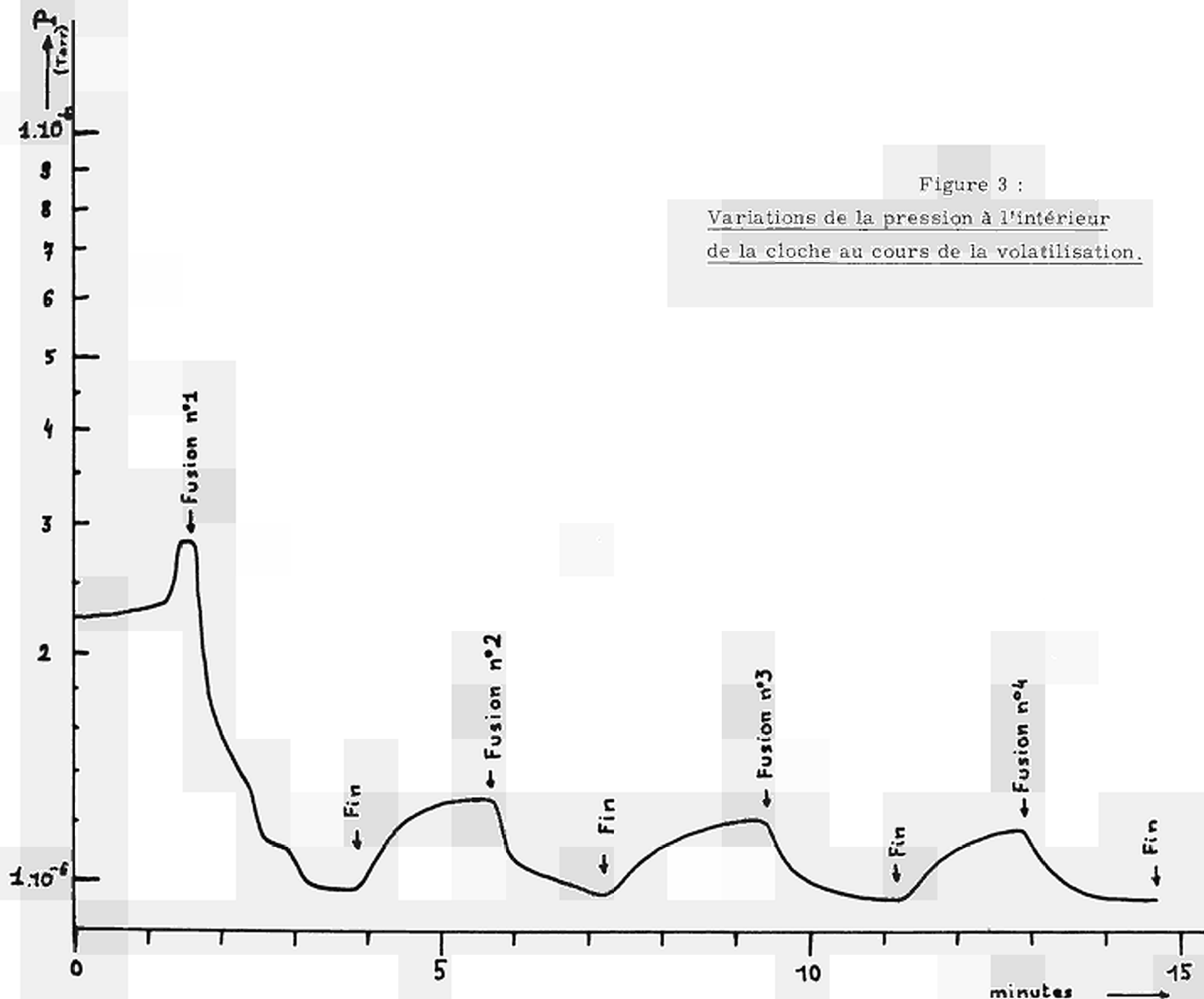
La pression à l'intérieur de la cloche est de 1 à $3 \cdot 10^{-6}$ Torr pendant la métallisation. Nous admettons que le titane n'est souillé par les gaz résiduels de la cloche que pendant le début de la volatilisation.

Nous avons essayé d'effacer le cache creuset plus tard, par exemple après que la distillation du premier creuset soit bien établie ; mais nous avons observé alors une remontée de pression importante dans la cloche, probablement parce que le dégazage du cache creuset lui-même est plus important que le pompage par le titane sur un trajet réduit.

Nous préférons augmenter la pureté du dépôt de titane en diminuant la distance des cibles aux creusets. On diminue ainsi le nombre de molécules de gaz résiduels rencontrées par le titane sur son trajet.

IMPREGNATION

Bien que l'imprégnation proprement dite ne dure que vingt minutes, l'opération complète est étalée sur 20 heures.



Chaque cible est placée dans un récipient étanche en pyrex ou en silice (figure 1) raccordé à la rampe à vide, et chauffé par un radiateur électrique (Photo rampe + radiateur, figure 4).

La cible à imprégner est au fond du récipient d'imprégnation. Le tube scellé, placé dans le récipient au dessus de la cible, sert à diminuer le volume total pour augmenter la sensibilité de la mesure de tritium.

Le récipient de stockage du tritium n'est pas représenté. Le radiateur a une puissance de 0 à 50 Watt, ajustable par un autotransformateur variable.

On vide l'appareillage jusqu'à 10^{-4} Torr. On prolonge le pompage une ou deux heures. On procède alors à un chauffage des supports, par paliers, jusqu'à 300°C. Chauffage et refroidissement, programmés électriquement, durent toute la nuit.

On isole alors le pompage. Le tritium est sorti par chauffage de l'uranium où il est adsorbé.

On mesure la pression de remplissage. On ferme les imprégneurs individuels et on procède à un nouveau chauffage; vers 250° à 300° C, température variable suivant l'épaisseur du titane, la nature et la forme des supports, l'imprégnation commence. On coupe le chauffage et le remplissage de la cible se poursuit pendant le refroidissement. Après lecture de la pression du gaz résiduel, on refixe le tritium dans l'uranium de son réservoir et on récupère la cible.

COMMENTAIRES

Cette méthode d'imprégnation a été mise au point empiriquement sur des installations données ; nous savons qu'elle ne peut pas être transposée directement sans déboires. Par exemple, nous avons observé quelquefois la fixation du tritium par le titane dès la température ordinaire, ce qui rendrait impossible sur nos installations la mesure de la quantité fixée.

Nous pensons, sans l'expliquer autrement, que notre recette de préparation des supports n'active pas le titane d'une manière propre à l'imprégnation à froid.

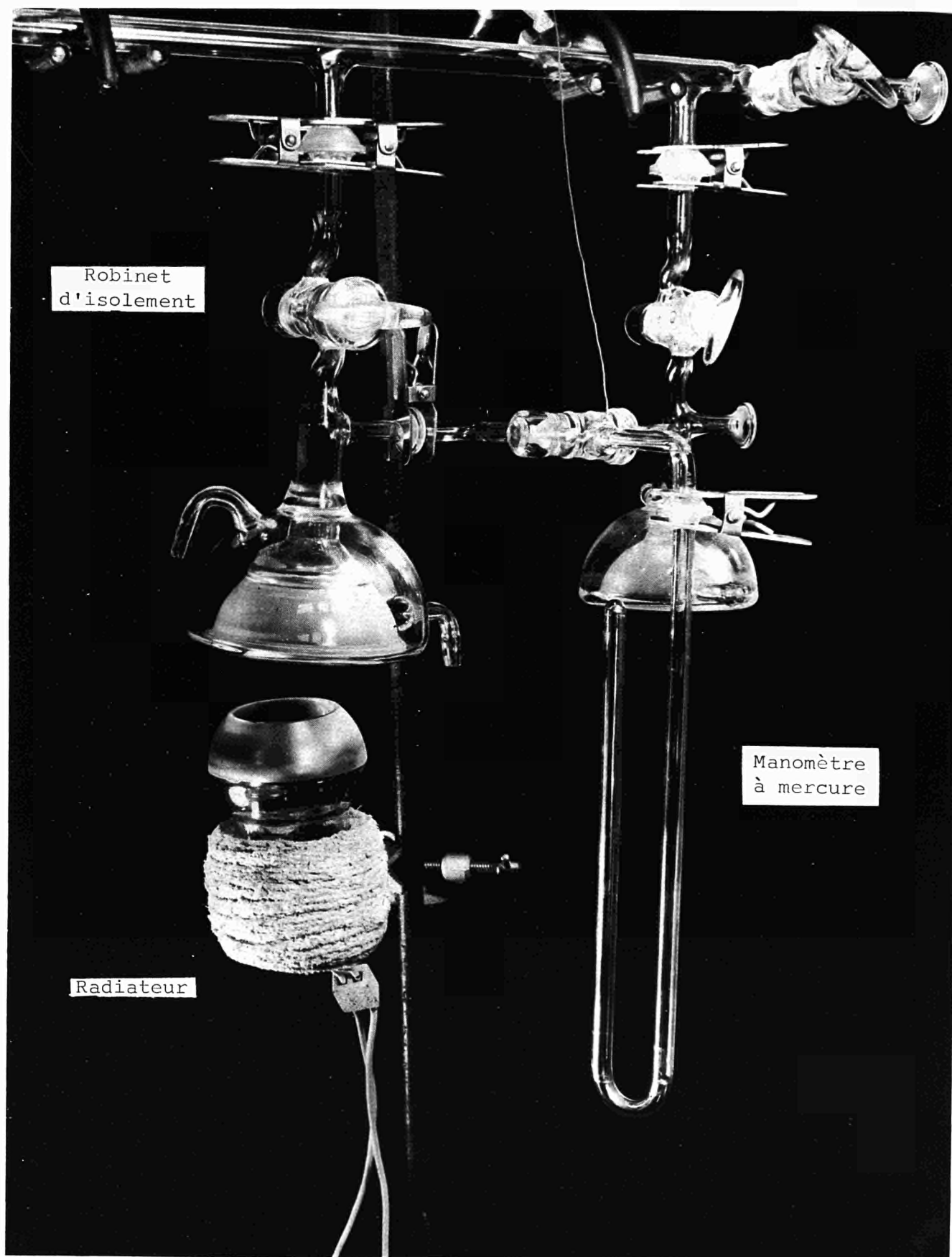


Figure 4 : rampe d'imprégnation
et radiateur de chauffage.

Nous avons également été gênés, dans l'élaboration de cibles spéciales, par la présence d'un manomètre à mercure sur le récipient d'imprégnation. Nous modifions alors l'appareillage pour isoler le manomètre, ou bien nous le supprimons.

Pour conclure, nous vous dirons quelque mots de nos possibilités :

- Nous avons fabriqué, ces dernières années, plusieurs milliers de cibles suivant ce mode opératoire.
- La fabrication normale est limitée aux cibles d'un diamètre de 49 mm. On peut fabriquer des cibles d'un plus grand diamètre, jusqu'à 150 millimètres mais le travail n'est pas un procédé de routine.
- En épaisseur de titane, nous nous limitons à 1 mg par cm², soit 2,5 microns épaisseur qui nous permet d'atteindre 1,2 curie de tritium par centimètre carré.

Les cibles plus épaisses se décollent et doivent être rebutées.

DISCUSSION

Mr. SMITH

I wish to confirm that you scratch the surface of your target backings in order to prevent flaking of the titanium and that you outgas the target at 300°C.

Mr. MANIN

Oui.

Mr. SMITH

We at Aldermaston manufacture our own target backings (Copper) which are highly polished. These are sent to Amersham. By degasing the backings nearer 600°C and deposition of the titanium at a high temperature little or no flaking occurs even at high ion currents on the targets. (> 100 watts/cm²). The titanium thickness is usually 2,5 mg/cm².

DERNIERS DEVELOPPEMENTS DANS LA PREPARATION DE
CIBLES TRITIEES A L'UNIVERSITE DE LIEGE

J.M. PETERS

Laboratoire d'Application des Radioéléments - Université de Liège -

LATEST DEVELOPMENTS IN THE PREPARATION OF TRITIATED TARGETS AT
LIEGE UNIVERSITY

ABSTRACT

Since the last Grenoble Conference, research on tritiated targets has been concerned firstly with the study and adaption as a tritium target of an organic polymer of high specific activity, synthesized from gaseous tritium in the most favourable conditions. The autoradiolysis of the tritiated compound is limited to an extremely low level. The calculated lifetime is 10^5 hours.

Secondly, research has been conducted into the optimum conditions for the fabrication of tritiated titanium targets. Various of metalization and impregnation techniques have been tested and compared.

The neutron yields obtained reach equivalent values for tritium contents lower than those of commercial targets. The half-lives are attained for a total cumulated charge of 3.2 to 3.4 Cb.

Research performed under contract no. 050-63-8 RISB and 095-64-11 RISB with the European Atomic Energy Community (EURATOM).

Les recherches entreprises à l'Université de Liège sur les cibles tritiées ont suivi depuis quelques 3 ans simultanément deux voies parallèles mais différentes.

La première consistait à expérimenter pour la première fois un matériau organique thermostable de haute activité spécifique comme cible tritiée, à savoir le polyphényle.

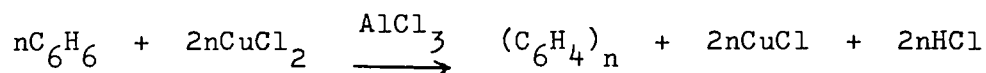
La deuxième voie plus récente avait pour but de rechercher les conditions optima de préparation de cibles de Titane et de les expérimenter par la suite sur d'autres hydrures métalliques.

C'est l'état actuel des travaux sur notre tentative de fabrication d'une cible plastique et les cibles classiques Ti que nous allons exposer.

Un projet de cible utilisant un matériau nouveau avait été présenté par M. GUILLAUME au dernier Congrès de Grenoble. Nous avons, à l'époque, simplement examiné une voie possible pour remédier aux inconvénients des cibles classiques. En effet, on supposait que si le tritium appartenait à une molécule organique stable, plutôt qu'à un hydrure métallique, sa stabilité s'en trouverait accrue, tout au moins dans les limites de stabilité thermique du matériau.

Le polyphényle qui venait d'être synthétisé par KOVACIC (1) semblait pouvoir répondre aux exigences qui étaient une stabilité thermique élevée alliée à une concentration d'hydrogène au moins égale à la concentration moyenne des cibles hydrures métalliques.

La littérature signalait qu'en présence de chlorure d'aluminium en tant que catalyseur et d'un oxydant cationique tel le chlorure cuivrique, le benzène se polymérise suivant la réaction



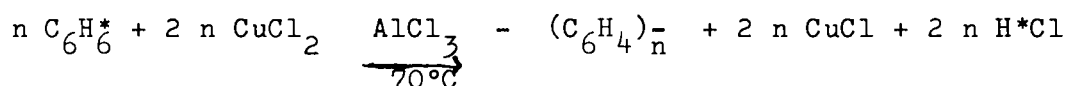
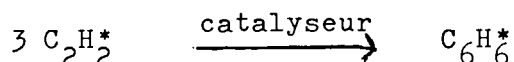
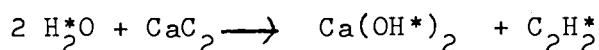
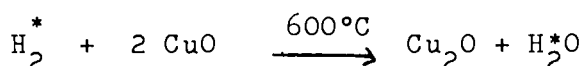
Les difficultés restaient nombreuses à surmonter pour obtenir du polyphényle tritié.

En effet, à côté des avantages que présente cette molécule, apparaissaient pas mal d'inconvénients dont les principaux étaient le très mauvais rendement chimique de la synthèse, l'insolubilité du polymère et l'exigence d'activité spécifique élevée qui écartaient le recours aux méthodes classiques de marquage par échange et imposaient un mode tout spécial de fabrication de la cible.

Les premiers résultats sur des cibles de polyphényle deutéré à 24%, rapportés au Congrès de Grenoble (65) nous avaient engagés à poursuivre la tentative.

Le principal problème était la synthèse de polyphényl tritié à haute activité spécifique. Celle-ci s'est avérée pleine d'obstacles. Ce sont les étapes de cette synthèse que nous allons exposer.

Etant donné l'inefficacité des méthodes de marquage par échange pour obtenir les activités spécifiques exigées la méthode de synthèse directe s'imposait à savoir



L'exposé de ce programme réclame un certain nombres de commentaires.

Il faut avoir à l'esprit que toutes ces synthèses doivent être microchimiques, qu'elles doivent s'effectuer à la chaîne en enceinte fermée avec des rendements à chaque étape, les plus proches de 100%.

Dans cette optique, la phase de formation de l'eau n'offre aucune complication. Par contre, la seconde équation montre que 50% de l'hydrogène de départ échappe dans l'hydroxyde de calcium.

La troisième réaction utilise un catalyseur de polymérisation de l'acétylène en benzène qui consiste en une alumine activée par des ions de valence élevée. On obtient des rendements compris entre 95 et 98% par l'utilisation d'alumine activée au Vanadium.

Quant à la dernière étape, nous avons dû l'adapter en microchimie et relever le rendement de 6% atteint par le procédé initial à 100% que nous obtenons actuellement.

La figure n° 1 montre la rampe de synthèse directe du benzène. Chaque partie de cette rampe a posé des problèmes particuliers très longs à résoudre.

Improvisation: (cf dessin)

- Détermination des débits gazeux, grandeurs des volumes, températures de réaction de synthèse de l'eau pour l'obtention d'un rendement maximum.
- Mise au point d'une technique de synthèse de l'acétylène récupérant l'eau retenue dans l'hydroxyde de Ca.
- Dans ce but recherche d'une préparation de carbure de Ca de haute pureté.

Des difficultés sérieuses également ont surgi lors de la polymérisation du benzène deutéré. En effet, des échanges très importants ont lieu au sein même de la masse réactionnelle. Nous avons cependant remédié à cet inconvénient par une série de précautions tendant à éliminer toute trace d'eau en cours de synthèse tant au niveau des réactifs que des volumes de réaction.

Nous pouvons de cette manière synthétiser environ 100 mg de polyphényl deutéré avec un rendement total calculé sur le deutérium mis en oeuvre au départ, supérieur à 80%, la pureté isotopique en Deutérium est supérieure à 85%.

Une dernière difficulté à résoudre était l'évacuation de l'acide chlorhydrique du volume de synthèse du polyphényl après réaction. Cette opération n'est évidemment nécessaire que dans le cas de synthèse de polyphényl tritié.

En faisant réagir l'acide chlorhydrique sur la vapeur de zinc à 900°C on dégage quantitativement le tritium lequel peut alors être stocké soit sur carbone actif soit sur four U.P.

Rappelons que la technique de fabrication de la cible consiste en une distribution homogène de poudre de polyphényl sur un disque d'acier préalablement traité par une laque d'argent, le tout comprimé sous 15 t/cm^2 et cuit au four à 300°C afin d'assurer l'adhérence par l'intermédiaire de l'argent.

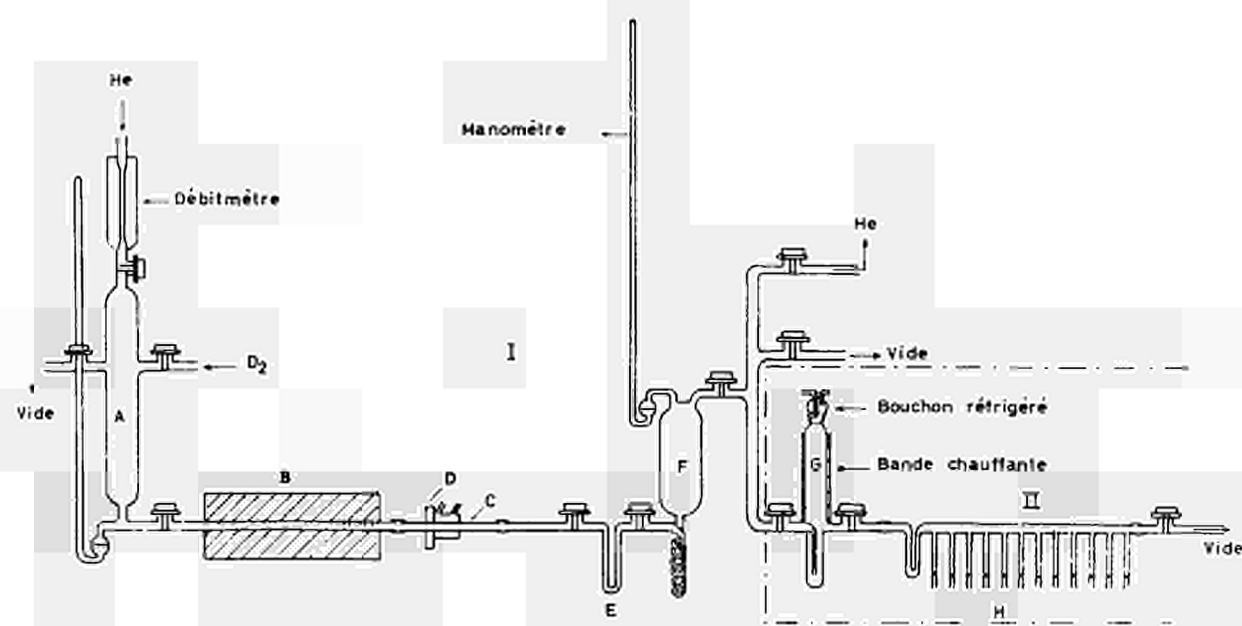


FIG. 1

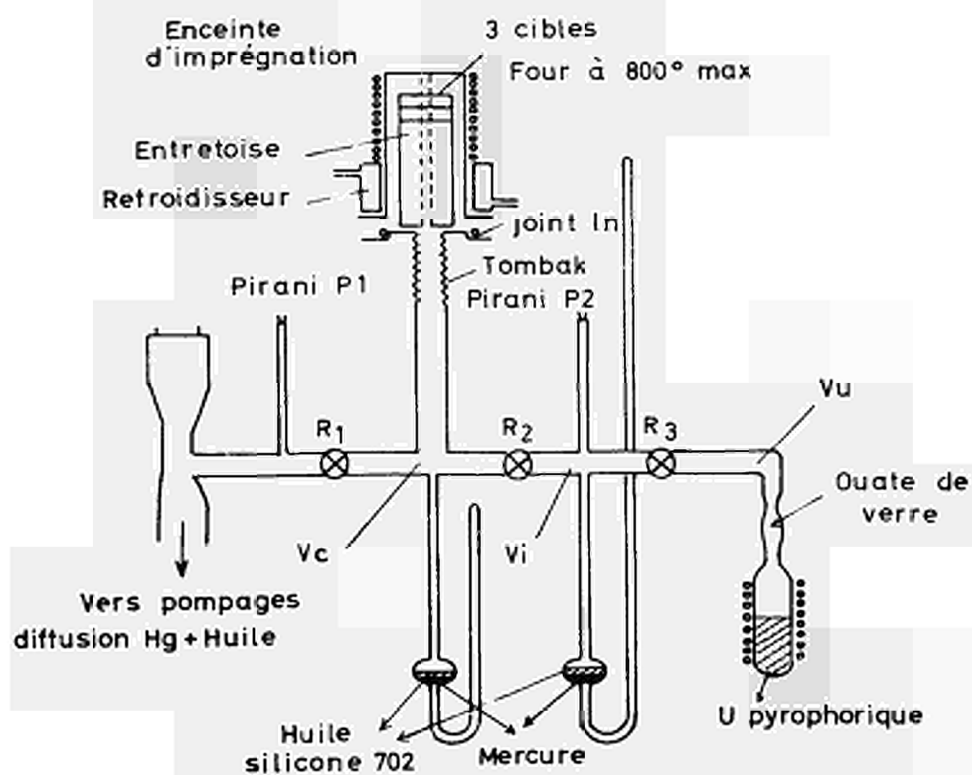


FIG. 2

Nous venons de préparer du polyphényl tritié à partir d'un mélange tritium-hydrogène dans la proportion volumique 1/4.

L'activité spécifique calculée, compte tenu des rendements isotopiques de chaque étape devrait être à peu près de 10 curies par milimôle de PP.

Il ne semble manifester aucune tendance importante à une radio-lyse destructive, toutefois il apparaît à froid et à chaud un dégagement continu constant de tritium qui équivaut à 30-50 $\mu\text{C}/8\text{heures}$.

Par souci d'objectivité, nous parlerons des essais de cibles de PP sous accélérateur. Nous ne pouvons prétendre au caractère absolu et définitif de ces résultats. Nous ne possédons en effet aucune courbe définitive, étant donné la détérioration du PP dès l'instant où le courant de cible dépasse 120 μA .

A 100 KeV et des courant inférieurs à 100 μA les résultats présentés à Grenoble se confirment, à savoir qu'à 10% près on rejoint la moyenne des cibles classiques. Pour des énergies plus élevées, les cibles testées dégazent exagérément dès les premières secondes. Pendant les premières minutes, il n'est guère possible de stabiliser ni la tension ni le courant de telle sorte qu'aucune mesure de rendement neutrons ne peut être réalisée. La cible continue à dégazer ^{plus} faiblement pendant environ 1/2 h, après quoi le flux se stabilise à une valeur de l'ordre de 50% de celle de cibles de référence sans aucune décroissance appréciable même après plus de 10 h de bombardement.

Dans ces conditions, nous n'avons pu tester le polyphényl tritié bien que nous en ayons préparé dans ce but.

La surface des cibles, après irradiation, n'est pas uniformément détériorée. A côté de plages décollées et complètement carbonisées, d'autres n'ont subi aucun dégât, ce qui indique que le procédé mécanique de la fabrication des cibles nécessite encore quelques améliorations.

On peut d'ailleurs avec quelque habitude repérer avant les tests par un examen sous microscope, les plages qui subiront des dégâts.

Le problème majeur consiste à assurer à la pastille de PP une évacuation rapide des calories de surface. Le dernier essai en ce sens fut le test d'une cible de PPH contenant 15% de bronze d'Ag et recouvert d'un mince film métallique de 50 $\mu\text{g}/\text{cm}^2$. Celle ci n'a subi aucun dommage ni dégazage sous faisceau de 300 KV et 160 μA .

Nous sommes absolument certains, qu'en supprimant les causes de dégazage de nos cibles, leur rendement neutronique sera appréciablement augmenté.

Il reste acquis qu'une utilisation aux bas courants des cibles de PP assurera une stabilité remarquable du flux neutronique dans le temps.

SUR LES CIBLES METALLIQUES Ti-T

Nous avons d'autre part entrepris l'étude des conditions optima de préparation de cibles métalliques de titane.

Le but de cette étude préliminaire était d'estimer l'importance des facteurs physiques qui, au niveau des étapes de métallisation et d'imprégnation, conditionnent le rapport atomique H/Ti ainsi que le comportement de la cible sous bombardement.

Nous nous sommes inspirés des procédés les plus favorables renseignés dans la littérature et nous avons recherché les principaux facteurs conditionnant une fabrication reproductible et contrôlée des cibles de Ti, et susceptibles d'améliorer leur temps de vie.

Nous avons testé successivement les divers facteurs suivants :

1° Choix et traitement du backing

- Nature et pureté

Nous n'avons jamais pu mettre en évidence au cours de nos essais une quelconque influence spécifique de la nature ou de la pureté du backing sur la reproductibilité des propriétés des cibles hydrogénées. Les essais ont porté sur des backings de Cu, Ag et Au.

- Traitement avant métallisation

Il est apparu, avec évidence, que l'état de surface du backing métallique avant la métallisation avait la plus grande importance.

C'est cet état de surface qui, dans nos expériences, a conditionné, l'adhérence de la couche métallisée après métallisation et après imprégnation. L'adhérence sera d'autant moins bonne que la surface du backing sera polie.

Le traitement que nous faisons subir aux backings avant métallisation consiste en un sablage sec effectué dans des conditions rigoureuses.

Ces conditions de sablage n'ont été découvertes qu'après de nombreux essais réalisés dans différentes conditions.

Le sablage sec effectué est le seul qui conduise sans exception aucune à des résultats entièrement satisfaisants.

Remarque:

Différents auteurs prétendent que le traitement de surface préalable à la métallisation ne constitue pas une opération critique: la nature de ce traitement n'aurait aucune influence sur les qualités des cibles.

Nos expériences nous ont conduits à des conclusions tout à fait opposées.

Nous avons en effet testé successivement les traitements de surface suivants sur Ag et Cu, toute autre condition restant constante.

- polissage mécanique (brillant)
- décapage chimique
- dépolissage papier émeri
- dépolissage rubbing compound "Dupont"
- dépolissage abrasif SiCgrain
- sablage humide à $2,5 \text{ Kg/cm}^2$ - 4 Kg/cm^2 - 6 Kg/cm^2
- sablage sec à 400 gr/cm^2

Seul le sablage sec strictement opéré dans nos conditions a donné des résultats qualitativement et quantitativement positifs. Aucune cible provenant de ce mode opératoire n'a dû être rejetée au cours des nombreux essais effectués.

Après sablage, les surfaces sont lavées dans les vapeurs d'alcool isopropylique.

L'alcool isopropylique convient particulièrement bien étant donné son haut pouvoir de solubilisation.

2° Condition de métallisation

Le principal facteur conditionnant le succès de cette opération est la pureté de la couche métallisée.

Dans ce but, il importe:

- 1°) d'utiliser un métal à évaporer suffisamment pur

Dans nos expériences, nous avons utilisé du Ti d'origine très diverses mais toujours de haute pureté:

Titane Kroll T35

Titane préparé par électrolyse (C.N.R.S.)

Nous n'avons enregistré aucune différence dans les propriétés des couches métallisées au départ de ces deux seules qualités de Ti.

En ce qui concerne le chauffage des backings pendant la métallisation:

Divers auteurs insistent sur l'intérêt qu'il y a de chauffer les backings, au cours de la métallisation.

L'expérience nous a montré que ce facteur n'a aucune influence sur les qualités d'adhérence, ni sur la teneur en tritium de la cible préparée.

Il en résulte que nous effectuons désormais toutes les métallisations sur backings à température ordinaire.

3° Conditions d'Imprégnation

Le schéma général de l'imprégnation est représenté à la figure 2 .

Les principales améliorations d'un tel montage sont :

- 1) la limite du vide de dégazage avant imprégnation
- 2) l'absence de toute vapeur (Hg graisse) considérée comme poison éventuel de la surface métallisée.

Le mode opératoire d'une imprégnation s'établit dans les conditions actuelles, comme suit :

le vide est réalisé progressivement sur tout le montage, au moyen de deux pompes à diffusion en série.

Le dégazage des parois intérieures de l'enceinte d'imprégnation est poursuivi à froid jusqu'à un vide limite de $2 \cdot 10^{-7}$ torr et une remontée de l'ordre de 10^{-7} torr.litre.sec⁻¹.

L'enceinte d'imprégnation chargée des cibles est portée progressivement à température croissante jusqu'à 400-450°C.

La pression est constamment suivie en fonction du temps. Lorsque, pour une pression de $5 \cdot 10^{-7}$ torr, la remontée ne dépasse pas $5 \cdot 10^{-7}$ torr.litre.sec⁻¹, on procède à l'imprégnation.

L'étude de cette étape nous permet d'affirmer que

- 1) Une température de 400°C maintenue au niveau des cibles et sous un vide de $8 \cdot 10^{-8}$ torr à 10^{-7} torr conduit à des résultats très positifs. Des températures supérieures n'ont présenté aucune influence sur ces résultats.
- 2) Par contre, la vitesse d'introduction de l' H_2 au niveau des cibles chauffées doit être aussi faible que possible. La réaction d'absorption de H_2 par le métal est en effet d'autant plus grande que la pression partielle en H_2 est grande.
- 3) Afin de modérer la vitesse de réaction de l' H_2 sur le métal lors du refroidissement de la couche métallique, il est conseillé de contrôler la vitesse de ce refroidissement.
- 4) Les conditions de stockage entre la métallisation et l'imprégnation de la couche métallisée, influencent sensiblement le rapport atomique H/Ti de la cible.

Nous avons en effet montré que une durée minimum de stockage sous vide sec était la solution préférable.

Des pressions partielles croissantes en O_2 ajoutées dans le volume de stockage de même que des durées de stockage croissantes sous vide, ont conduit, toutes autres conditions égales, à des rapports H/Ti décroissant linéairement.

RENDEMENT NEUTRONS

Les tests de cibles tritiées préparées dans ces conditions ont été effectués au moyen de notre accélérateur Van de Graaff 400 KeV. Les rendements en neutrons et les demi-vies ont été déterminés pour des durées de fonctionnement continu atteignant 6 à 7 heures.

Le faisceau de deutons n'est pas défléchi, il n'y a donc pas séparation des ions atomiques et moléculaires. L'homogénéité du faisceau est préalablement déterminée sur lame de quartz tandis que son étendue est estimée par l'intermédiaire d'un collimateur.

Le porte cible assure un refroidissement d'eau de l'arrière de la cible.

L'étanchéité au vide est assurée par un joint O-Ring, placé à l'arrière de la cible.

Le flux de neutrons instantané est mesuré par un long compteur BF_3 entouré de paraffine et d'une feuille de 4 mm de Cd.

L'irradiation de disques de Cu suivant la convention du Texas permet la conversion de la réponse BF_3 en rendement neutrons par sec.

L'analyse du graphique n° 3 conduit aux constatations suivantes:

1. Les débits neutrons sont au début de chaque essai du même ordre de grandeur que les cibles commerciales soit de 1 à $4 \cdot 10^8$ n/ μCb , bien qu'obtenus au moyen de cibles dont la teneur en tritium ne dépassait pas 1 Curie.
2. Les décroissances jusqu'à 150 μA suivent une courbe qui ne présentent pas l'allure pseudo-exponentielle bien connue.
3. Des cibles fabriquées dans les mêmes conditions d'imprégnation donnent, sous bombardement, des comportements parfaitement identiques.
4. Les premiers essais effectués tendent à prouver l'intérêt majeur qu'il y a d'une part de réduire au maximum l'épaisseur du backing et d'autre part de défocaliser le faisceau sur toute la surface utile de la cible.
5. Les différents essais que nous avons effectués nous indiquent que la 1/2 vie de nos cibles est actuellement atteinte pour une charge moyenne de 3,5 Cb.

Ces essais vont être systématiquement poursuivis afin de pouvoir dégager les facteurs qui en cours de fabrication conditionnent la tenue sous deutons des cibles métalliques.

C'est dans cette optique, que nous avons étudié la construction d'une cellule mixte de métallisation et d'imprégnation sous UHV.

Cette cellule mixte de métallisation et d'imprégnation - figure 4 - réalisée en verre pyrex sera adaptée à un groupe de pompage à ultra-vide constitué par un ensemble combiné: pompe ionique + pompe à sublimation de vitesse de pompage de 400 l/sec, et de vide limite inférieur à 10^{-10} torr.

Après la métallisation des 2 backings soumis à rotation continue (afin d'obtenir un dépôt homogène) ceux-ci seront transférés, à l'intérieur de la cellule, dans la zone d'imprégnation équipé d'un manomètre et de la source

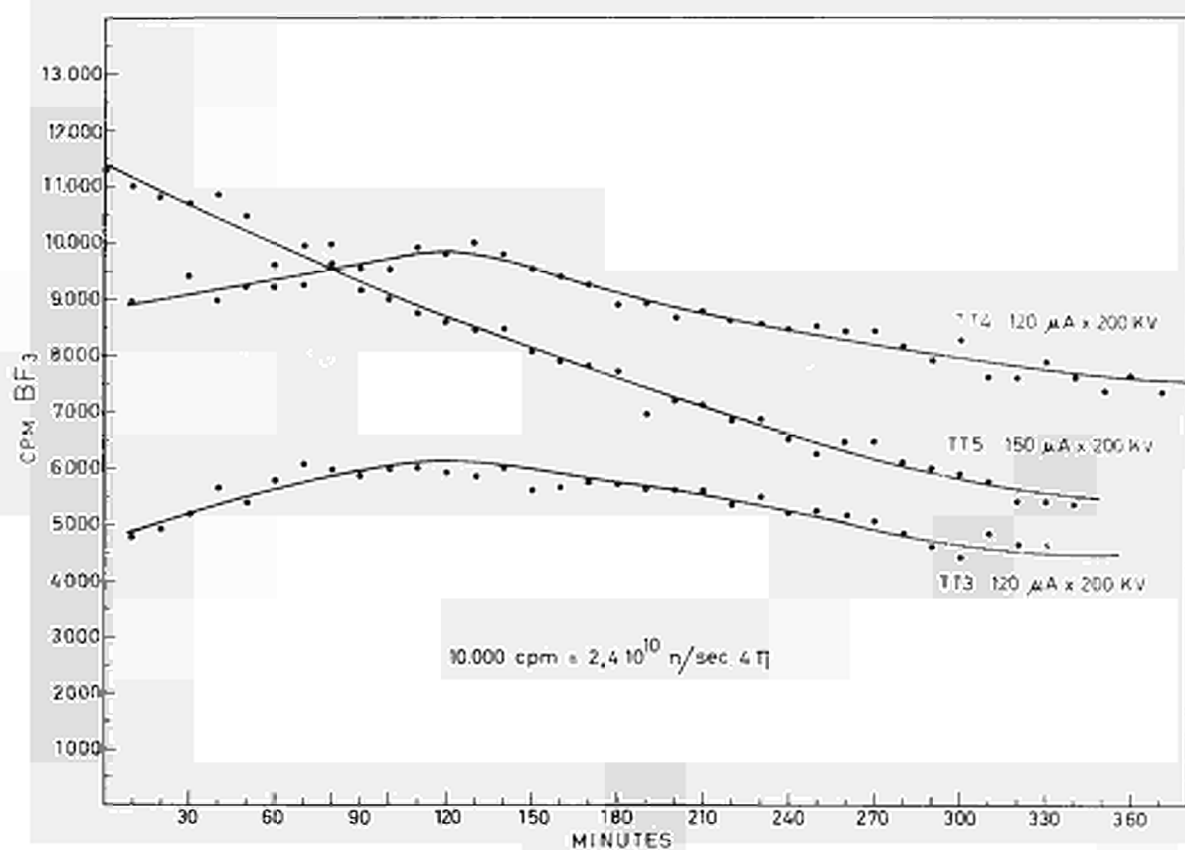


FIG. 3

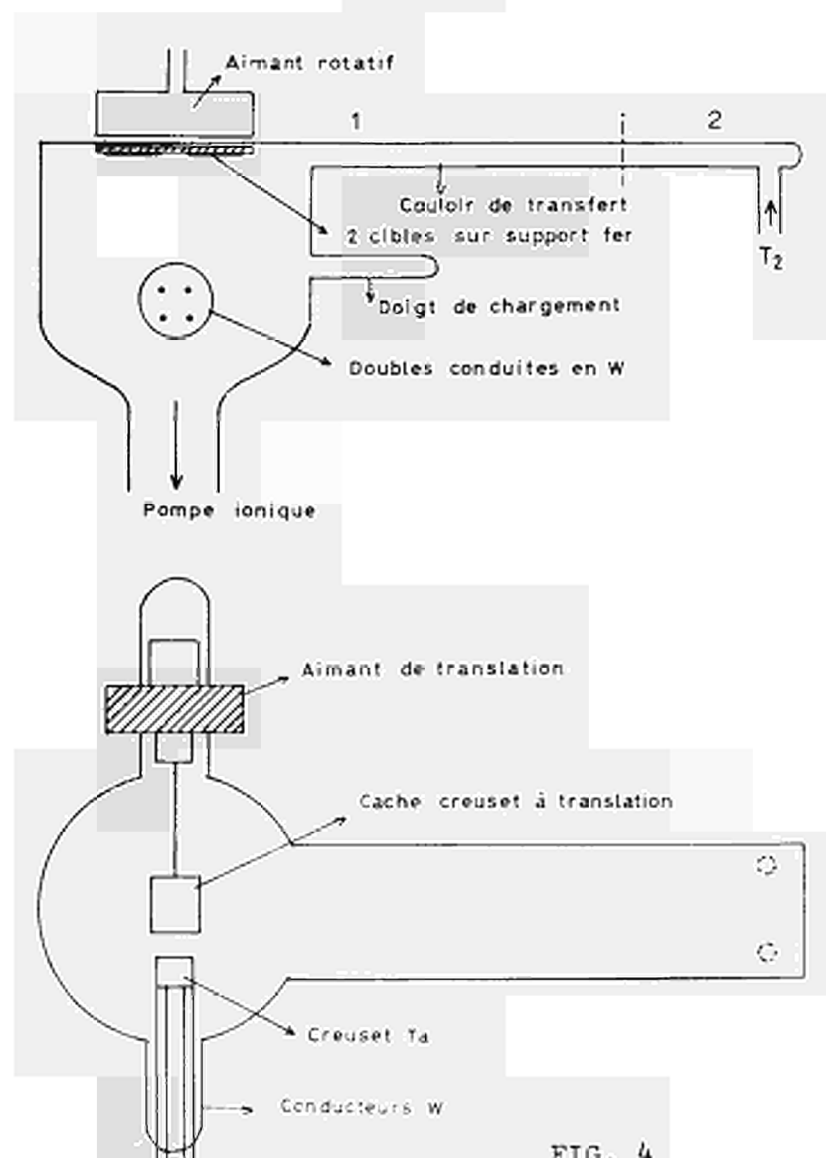


FIG. 4

tritium. Cette zone sera détachée du métalliseur proprement dit par scellage. L'imprégnation pourra donc s'effectuer directement après la métallisation sans retour à la pression atmosphérique.

La réalisation d'un tel projet nécessite des mises au point techniques délicates qui sont en cours d'étude actuellement.

DISCUSSION

Mr. MOUTHON

Dans le cas de vos cibles métalliques, quel est le rapport atomique $^3\text{H}/\text{Ti}$?

Etant donné le très bon vide sous lequel vous travaillez en imprégnation , n'avez-vous pas observé une imprégnation à froid des cibles ?

A Sodern, où nous travaillons en imprégnation avec un pompage ionique, l'imprégnation s'effectue à la température ordinaire, fait très remarquable.

Mr. PETERS

Les cibles métalliques avec lesquelles nous avons établi nos courbes de rendement avaient un rapport atomique $^3\text{H}/\text{Ti}$ compris entre 1,5 et 1,6.

Nous admettons toujours le tritium au niveau de cibles chauffées à 400°C. On contrôle à la fois la vitesse d'introduction de gaz et la vitesse de refroidissement de la cellule d'imprégnation.

Peut-être cependant Mr. GUILLAUME aurait-il un avis sur cette question étant donné qu'il s'occupe tout spécialement des cibles titane. Je ne crois pas qu'il ait des résultats quantitatifs.

Mr. REIFENSCHWEILER

Sind Werte über Ausbeute und Lebensdauer von tritiierten Polyphenyl-targets bekannt ?

Mr. PETERS

Non.

- 1) La surface utilisable de polyphényle a un diamètre de 17 mm. Ce choix a été fait pour des questions de commodité de distribution de la poudre de polyphényle sur le support.

Nous avons utilisé des faisceaux de 1 à 1,5 cm de diamètre. Cependant la plupart des essais ont été fait avec un impact de 1 cm de diamètre qui nous semblait une bonne valeur pour se faire une idée de la stabilité du matériau sous bombardement.

2) La poudre est distribuée uniformément sur la surface du support à recouvrir (12 à 15 mg de polyphényle).

Le support est préparé préalablement de la manière suivante :

- a) une couche de laque d'Ag cuite au four vers 350 - 400°C
- b) une seconde couche séchée une heure et non cuite

Le polyphényle est pastillé sous 15 T/cm²

La cible est cuite au four programmé :

- 3 heures de montée jusqu'à 300°C
- 3 heures de chauffée à 300°C
- 3 heures de refroidissement progressif.

Mr. MOUTHON

J'ai remarqué des divergences de point de vue entre votre exposé et celui de Mr. MANIN au sujet des conditions optimum de préparation des cibles (sablage sec) et de métallisation (température du support). Peut-on expliquer ces divergences d'une façon thermique où cela vient-il de conditions opératoires bien particulières. J'aimerais que Monsieur MANIN expose son point de vue.

Je suis d'accord pour reconnaître avec Mr. PETERS que l'adhésivité du titane dépend d'abord des conditions préparatoires du support.

Mr. PETERS

Pour nous la surface du backing métallique avant la métallisation a la plus grande importance.

Nous effectuons toujours un sablage sec dans des conditions rigoureuses. Tous les autres prétraitements de la surface des cibles que j'ai cités dans mon texte ont conduit à des résultats décevants.

Le chauffage des supports en cours de métallisation n'est pas nécessaire.

Mr. MOUTHON

Monsieur MANIN est d'accord avec vous pour reconnaître que l'imprégnation est meilleure lorsqu'on métallise à froid. Cependant dans ce cas, l'adhésivité du titane sur son support est mauvaise.

Avez-vous remarqué entre vos cibles sablées et des cibles polies des différences du point de vue émission secondaire et pulvérisation ?

Sinon, il est probable que la densité de courant étant bien plus faible dans vos expériences que dans le cas des expériences du CEA, les phénomènes de pulvérisation sont très faibles dans votre cas indépendamment de la qualité des cibles.

Mr. MANIN

Pour répondre aux divergences apparentes, que souligne Mr. MOUTHON, entre les conclusions de Mr. PETERS et de Mr. MANIN sur les causes de décollement de la couche de titane, nous proposons d'examiner successivement les 3 cas suivants :

- 1) le titane se décolle dès la métallisation : c'est un défaut grossier, qui entraîne le rejet de la cible; ce défaut peut provenir : d'un nettoyage insuffisant - d'un mauvais vide dans la cloche - ou n'importe quelle autre cause.
- 2) le titane se décolle au moment de l'imprégnation : le décollement peut être causé a) par des impuretés, ou encore pour des raisons
b) physico-chimiques : gonflement exagéré du réseau cristallin du titane lorsque le taux de remplissage dépasse 2 ; ou encore pour
c) des raisons inconnues.
- 3) le décollement se manifeste sous accélérateur. En plus de toutes les raisons précitées qui peuvent le provoquer, avec retard, il peut être provoqué par la pulvérisation du titane sous l'effet des deutons.

Il ne faut donc pas s'étonner que l'on puisse accroître l'adhérence du titane

- dans certains cas en polissant le support
- dans d'autres cas en le dépolissant - (sablage ou grattage)

Mr. ROCHE

- I) Quelle est la stabilité dans le temps des cibles au polyphényle tritié sous l'effet de la radiolyse induite par les β du tritium ?
- II) N'y a-t-il pas des problèmes de mesure du courant cible avec une cible plastique qui est vraisemblablement isolante ?

Mr. GUILLAUME

- 1.) Les premières mesures de la radiolyse du polyphényle ont donné 40 à 50 $\mu\text{C}/8\text{h}$ de tritium pour 12 mgr de produit intervenant dans la fabrication d'une cible. Cela conduit à une vie de l'ordre de 10^5 heures, pour une cible plastique.
- 2.) Non, il n'y a aucun problème de mesure de courant, même sur surface isolante comme une cible de polyphényle. La mesure s'effectue tout à fait normalement. Cependant, pour **amener** une meilleure conductivité et par là, une température de surface plus faible, nous métallisons la surface du polymère.
La mesure du courant est rendue difficile uniquement à cause du phénomène de dégazage des points chauds de surface qui apparaissent dès le début de l'irradiation sous deutons, d'où pour nous, l'obligation d'amener une meilleure adhérence par une meilleure répartition du polyphényle en surface du backing.

Mr. SMITH

- 1) The life of your Ti.T. Targets of approximately 3.3 coulomb corresponds with 1 mA hour.
- 2) Sanding the targets leaves a rough surface, when titanium is deposited on a rough surface it is likely to deposit preferentially on the tops of any high spots or is liable not to deposit so well in the hollows and poor heat conduction may occur in these hollows resulting in reduced life.

Mr. GUILLAUME

- 1) La demi-vie de nos cibles de titane est pour 150 μA sous 250 kV de 6 à 7 heures soit de 3,2 clb pour une surface d'environ 1 cm^2 , soit encore comme vous le dites de 1mA heure.
- 2) Il apparait, à l'examen sous microscope, que le titane métallisé recouvre parfaitement la surface traitée du backing en épousant les irrégularités. Soumise à des chocs thermiques répétés la couche métallisée n'a jamais craquelé, dans ces conditions.

Une interprétation peut être au contraire avancée suivant laquelle, étant donné la rugosité de la couche de titane, la surface efficace qui participe au transfert de chaleur est plus grande que la surface plane correspondante. Il en découlerait des températures superficielles locales plus faibles que sur backing poli.

Mr. REIFENSCHWEILER

Zu dem Problem einer Beschränkung der Lebensdauer der Targets bzw. einer Verkürzung der Halbwertsdauer der Targets durch Abdampfen des Titans von den durch das Sandstrahlen verursachten Erhöhungen möchte ich folgendes ausführen.

Unsere in abgeschmolzenen Neutronenröhren verwendeten Targets wurden ebenfalls vor dem Aufdampfen des Titans durch trockenes Sandstrahlen behandelt. Da wir die Technik der Selbsttargets mit gemischtem Deuterium-Tritium-Ionenstrahl anwenden, tritt keine Abnahme der Neutronenausbeute durch Austreiben des Tritiums aus dem Titan ein. Die Lebensdauer ist vielmehr durch das Zerstäuben oder ein eventuelles Verdampfen des Titans gegeben, und der oben beschriebene Effekt müsste deshalb deutlich wahrnehmbar sein. Bei Lebensdauerexperimenten bis zu $600 \text{ mA} \times \text{h}$ mit einer Belastung von $100 \text{ mA} \times \text{h/cm}^2$ (Targetoberfläche 6 cm^2) wurde jedoch keinerlei Abnahme der Neutronenausbeute festgestellt, es trat also kein Verdampfen des Titans von den Erhöhungen auf. Der Targetstrom betrug hierbei bei einigen Experimenten 100 oder 200 μA bei einer Beschleunigungsspannung von 125 kV, bei anderen Experimenten zwischen 2 und 5 mA bei einer Beschleunigungsspannung von 150 kV.

Mme. BREYNAT

Présentation de 2 photos montrant des cibles après irradiations

l'une dont le support est poli

l'autre dont le support est volontairement dépoli

conditions d'irradiations : 1 mA sous 300 keV $\varnothing = 26 \text{ mm}$

faisceau homogène

durée d'irradiation entre 6 et 7 heures.

Mr. DETAIN

Au sujet des évaluations du sputtering des cibles je peux préciser que nous avons fait quelques mesures concernant l'importance de ce phénomène pour des cibles titane. Nous avons trouvé des valeurs de perte de poids, de l'ordre de 10^{-2} mg/cm²/coulomb pour des cibles de diamètre de 49 mm bombardées par des deutons de 300 keV avec un faisceau de 1 mA, de diamètre de 25 mm. La température des cibles pendant ces expériences était de 100°C.

Mr. FABIAN

Wir haben, um die Haftfestigkeit von aufgedampften Titanschichten zu untersuchen, auf verschieden präparierte Cu-Unterlagen gedampft. Dabei konnten wir feststellen, dass das Titan auf sandgestrahlten geschmirgelten, geätzten und polierten Cu-Unterlagen gleich gut haftet. Das Abblättern schreiben wir einer unvollkommenen Reinigung zu.

Mr. REIFENSCHWEILER

Die Kurven von Fort und Huet über die Verteilung des Tritiums im Titan-target beweisen m.E., dass Titan und Kupfer an der Grenze der beiden Metalle ineinander diffundiert sind. Die Löslichkeit von Wasserstoffisotopen in Kupfer (als einem endothermen Wasserstoffabsorber) ist mehrere Größenordnungen kleiner als für Titan (als einem endothermen Wasserstoffabsorber). Die Tritiumkonzentration müsste also beim Uebergang von reinem Titan zu reinem Kupfer auf einen sehr kleinen Wert absinken. Der stetige Verlauf der Kurve an der Grenze mit einer ziemlich hohen Konzentration in der Kupferzone kann nur durch Anwesenheit von Titan in dieser Zone erklärt werden.

Mr. SMITH

Comment on distribution of tritium in titanium targets.

The last two speakers indicated that there is alloying of the titanium and copper at the interface. Such an alloy can presumably contain tritium and result in the tritium distributions shown. Work at A.W.R.E. by P.FIELDHOUSE, D.S. MATHON and E.R. CULLIFORD has tended to confirm this tritium distribution.

Complete removal of the titanium to leave only the copper backing results in very little tritium being present.

Use and possibilities of tritium and deuterium targets in a 150 keV pulsed ion accelerator at low average currents.

F. CORDERO

Junta de Energia Nuclear, Division de Fisica Madrid, Espana

ABSTRACT

We report some measurements concerning deuterium and tritium targets suitable for a low-energy accelerator (150 keV) to obtain a pulsed neutron source. The geometry and the operating conditions of the targets are described in connection with the study of the factors affecting the practical service life. Measurements of the rate of decrease in yield of neutrons from semi-thin tritium-titanium targets have been made. The yield falls rapidly to one-half when the integrated current reaches 0,7-1 coulombs (2.5 to 3 hours half-life at $50\mu\text{A}$); the neutron flux drops in about a factor of 10 after 5 C. No noticeable decrease in the target neutron yield is observed after ~ 50 C. By using deuterium titanium targets with contamination of oil vapors the neutron yield increases until reaching saturation with ~ 70 C (~ 12 C/cm²); subsequently, the yield remains constant. The processes which determine these results are discussed.

1. Introduction

We have at JEN (Junta de Energía Nuclear), Madrid, a low energy (150 keV) Cockcroft-Walton accelerator. It was built to provide a pulsed neutron source in a heavy water uranium subcritical assembly (1) for reactor physics studies. This machine has been described in (2) and experiments in (3). During the experiments we have made measurements of relative neutron yield. It is intended in this paper to report the results obtained with deuterium and tritium titanium targets. The purpose of our study is to define the performance possibilities of these targets in our accelerator with conventional vacuum system.

A neutron intensity of $10^8 - 10^9$ n/s is generally sufficient for many experiments. Using deuterium targets and an average current of several hundreds of microamperes, this accelerator may produce 10^8 n/s. With tritium targets it is possible to obtain 10^{10} n/s; in fact the yield is limited in practice by the strength of the targets under deuteron bombardment; a layer of carbon covers the targets and the neutron intensity that can be obtained from a given target is, for most of the time, much smaller than the value derived from calculation of the D-T reaction.

The duty ratios to be used must be low (2-5%); the pulse in intensities, however, must be high in order to perform measurements in reasonably short times.

2. Targets: mounting and cooling

Targets for this pulsed accelerator have to be used during long periods of time without much neutron intensity loss because it is not easy to change the target, due to very restricting space and geometry conditions. Figure 1 shows the output section of the accelerator. We place the target in the center of a large heavy water tank in order to inject neutrons into the subcritical assembly. Target extension tubes up to 2 meters in length have been used (~ 4 m from the ion source).

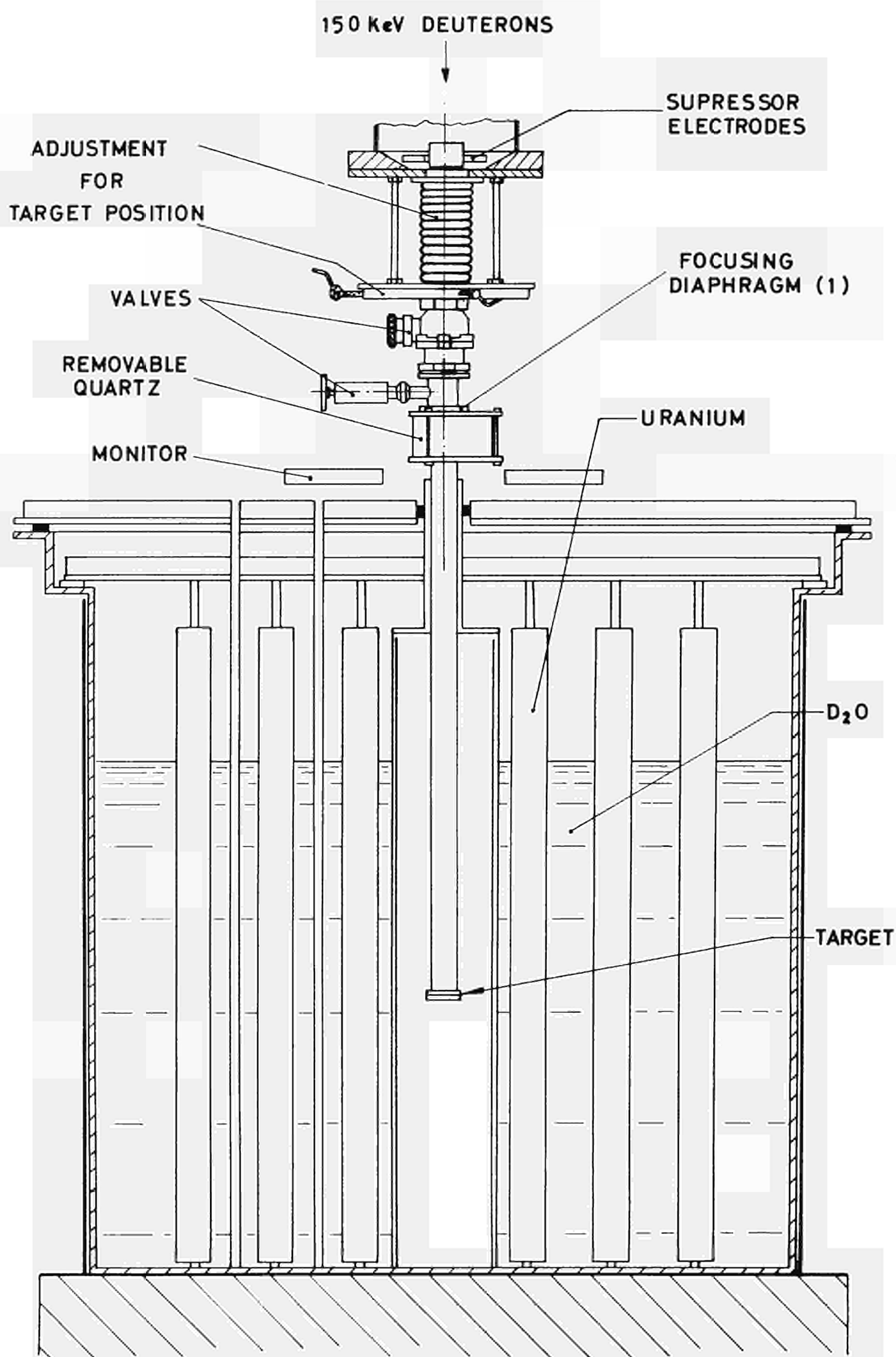


FIG.1.- TARGET EXTENSION TUBE IN
SUBCRITICAL ASSEMBLY

The use of a gas target is not possible due to the excessive energy loss of deuterons through the window. Consequently solid targets must be used. For these experiments heavy ice targets are not suitable because of their larger dimensions. The use of titanium targets simplifies the construction of the target holders and allows reduction of the dimensions to a minimum. We have employed standard deuterium and tritium titanium targets, 25 mm in diameter, supplied by Amersham. We use titanium targets because they give a somewhat higher neutron yield than the zirconium ones at the same beam current because of the higher atomic ratio per unit thickness (4). Table 1 summarizes data of targets that we have used. We have employed semi-thin tritium-titanium targets, about 1 μ thick, in order that the deuterons may terminate their paths further away from the active titanium layer and, in this way, increase the half life because the deuterium gas from the deuterons is not diffused back through the copper backing to the titanium layer (5). The deuterium titanium targets are thick.

Table I

TYPE	Ti weight [*] (mg)	Gas absorbed [*] (cm ³ N.T.P.)	Ti Thickness (μ g/cm ²)	Atomic ratio
CTB-664	2,5	0,67	390	1,46
CTB-666	2,6	0,59	410	1,00
CTB-669	1,8	0,72	280	1,73
DTB-5,561	6,8	1,6	1.330	1,01

Copper-backed targets have been soft soldered to a target holder without loss of tritium; this procedure gives good thermal conduction. The back side of the target was water-cooled; this cooling system is better than the annular cooling (6). Figure 2 shows the target holder.

The tritium and deuterium titanium targets have been used with sufficient cooling to maintain their temperature quite below 100° C.

* Data supplied by Amersham. Buckinghamshire, England

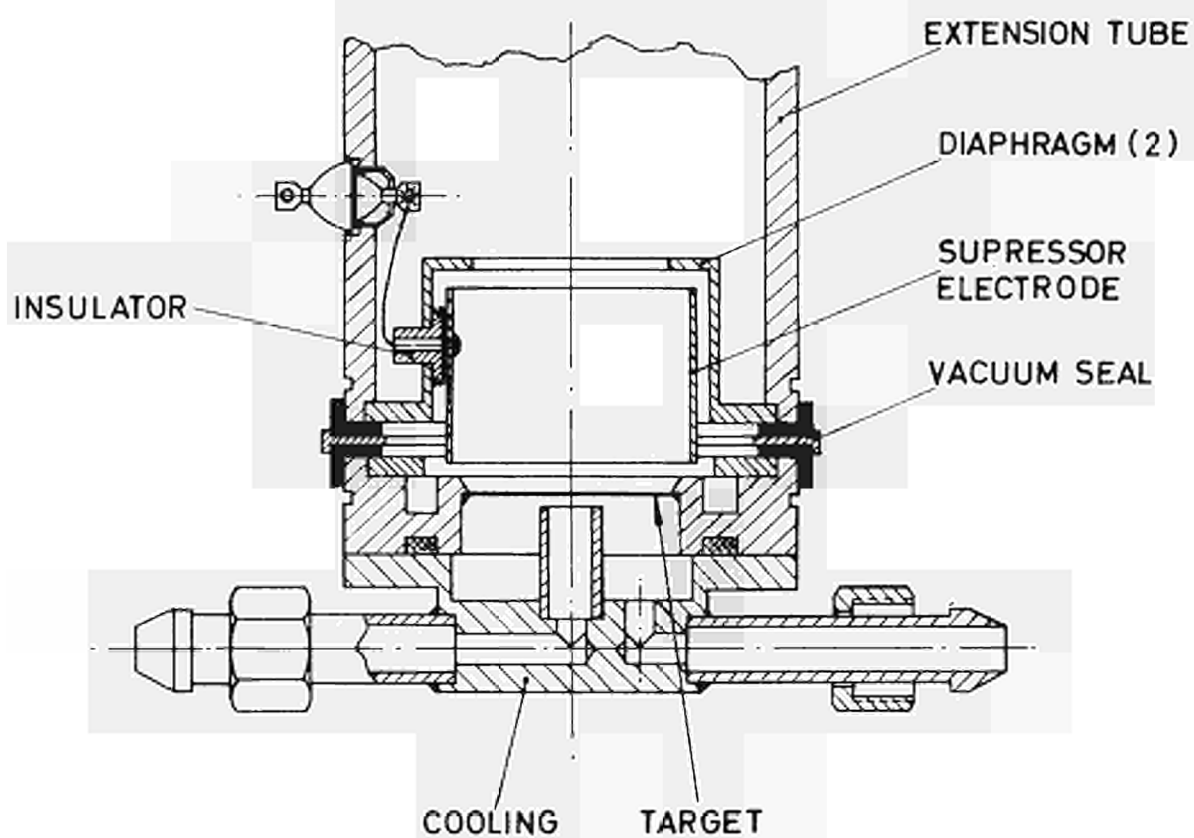


FIG. 2.- TARGET HOLDER WITH ARRANGEMENT FOR MEASURING ION CURRENT

Because of the ion bombardment there is a superficial heating effect of the target; with temperatures of the order of 100-200° C the tritium already escapes from the titanium (7) and this effect is one of the factors determining the target life. We have measured by means of a thermocouple the temperatures at different operating conditions in a copper disc of same dimensions as the targets employed (0,2 mm thick). With a voltage of 150 kV and an average current of the order of 500 μ A (8 mA peak current), i.e, 75 W for a diameter of 25 mm., we keep the target at a temperature below 50° C.

3. Operating conditions

3.1. Vacuum system.

A clean vacuum system is very important for preserving targets from hydrocarbons to increase their life; thus, the desirability of having such a vacuum system without organic materials. However, materials like neoprene o-rings, vacuum greases and oil for mechanical fore pumps and diffusion pumps are used. In order to prevent backstreaming of oil vapors from the diffusion pump, a water-cooled baffle and a refrigerated baffle (- 40° C) are used. The minimum pressure obtainable in the system is $\sim 10^{-6}$ Torr. The pressure during operation is in the region of $7 \cdot 10^{-6}$ - 10^{-5} Torr, depending on the ion beam current.

3.2. Ion beam.

Due to the fact that pulsed experiments are made with low duty ratio but high peak intensity and that the distance between electrostatic lens and target is large, one needs a wide and strongly convergent beam. Spherical aberration causes distortion of the image; thus, we observe that the spot (~ 1 cm diameter) is surrounded by a halo. The density distribution on the target is non-homogeneous.

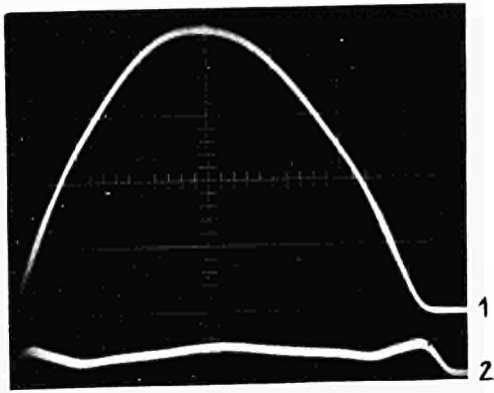
The beam is limited by diaphragms. A movable remote control quartz disc above the target provides a visual control of the beam focusing and centring (Fig. 1). The beam is focused to a diameter of approximately 25 mm at the target.

We use a Penning ion source; the amplitudes used were between 1-2 A peak current. The pulse generator and characteristics of the ion source are described in (8). The ratio of atomic to molecular ions is estimated to be about 30%; this ratio improves with current density in the ion source. Experiments with tritium targets have been made with pulses of sinusoidal form (Fig. 3 A). The current pulses were rectangular (Fig. 3 B) during the use of deuterium targets. Measurements have been made with the accelerator operating at 145-150 kV and 3-9 mA peak current; average currents from 30 to 400 μ A, duty ratio of 2-5%, repetition rate of 10-50 p.p.s and pulse length of 0,5-3 msec.

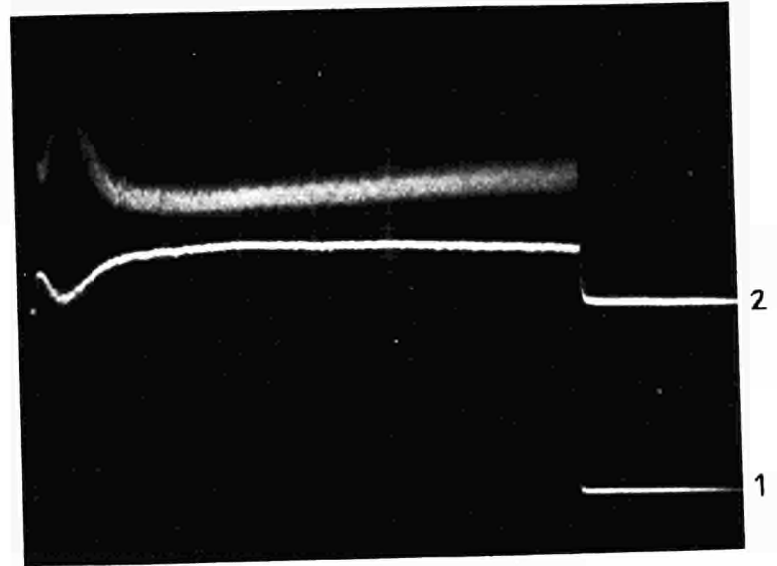
The beam current was measured at different electrodes of the target extension tube (Fig. 1 and 2). The opening in the suppressor electrode is larger than that in the diaphragm D_2 , so that no part of the direct ion beam can strike that electrode. The suppressor electrode was held at negative potential of - 1000 V, in this way, electrons coming down the accelerator tube (e.g. secondaries from the unfocused beam striking the extension tube) and secondary electrons produced in the target could not affect the measurements of the incident ion current on the target. Results of beam intensities versus the suppressor electrode bias are shown in Fig. 4. It is necessary to use a suppression voltage above 800 V; this value is higher than usual ones (9), probably, because an increase in pressure associated with pulsed operation causes a number of secondary processes (10).

3.3. Monitors

The neutron flux was measured by two counting channels located at the top of the subcritical assembly (Fig. 1). The counters have been placed at a distance 1-3 meters from the target at about 90° to the beam axis. The formation of self-loading targets on diaphragms 1,2 creates an additional D-D neutron background on the monitors; this cause gives the largest dispersion of average values ($\sim 20\%$). We have used stilbene and Hornyak scintillation counters and BF_3 gas counters.



A. SINUSOIDAL



B. RECTANGULAR

1) TARGET I: 0,5 mA/cm; T: 0,5 msec/cm
2) DIAPHRAGM (1); I: 0,2 mA/cm

1) TARGET I: 2,5 mA/cm; T: 0,2 msec/cm
2) DIAPHRAGM (1) I: 0,2 mA/cm

FIG. 3 CURRENT PULSES IN THE TARGET

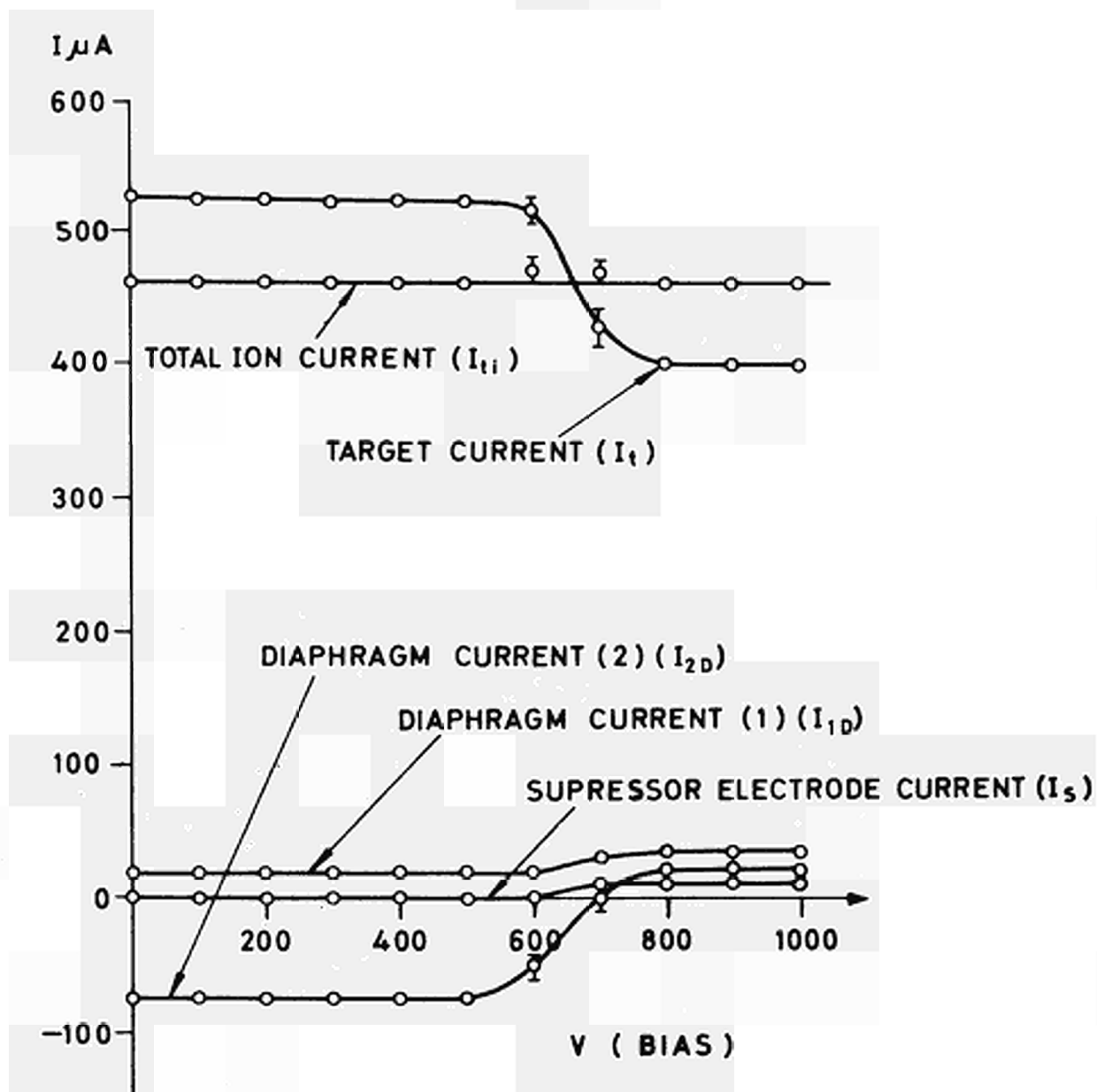


FIG. 4.- EFFECT OF THE SUPPRESSOR ELECTRODE BIAS VOLTAGE ON TARGET ASSEMBLY CURRENTS

$$I_{ti} = I_t + I_{1D} + I_{2D} + I_s$$

4. Results and discussion

The neutron yield changes of titanium targets have been examined; long-term and short-term tests have been made.

Figure 5 shows the performance of the tritium targets and the evolution of the neutron yield per microcoulomb as a function of the charge received; half lives are increased by about the ratio of the beam currents (4,9), because of that we integrated the ionic charge. These curves were obtained at 30-200 μ A average currents and 150 kV. Generally previously published results (5,7,11,12) cover lower loads. Relative neutron yield is in arbitrary units and the curves are separated so that the half lives can be seen. In the first portion there is a tendency in the half life to increase with the target activity in agreement with (13). There seems to be a two component time decay of the neutron yield; first, a fast drop in the yield is produced because organic vapors form under ion bombardment a carbonaceous coating on the target; subsequently there is a slow fall, probably because the formation rate of the contamination layer is slower, due to the surface temperature increase; at above 80-100° C the carbon deposition is smaller (4). Also processes of titanium degassing and displacement of the tritium by deuterium are possible. We have half lives of 2,5 to 3 hours at 50 μ A (1st zone). The neutron yield drops in about a factor of 10 after 5 coulombs and no noticeable decrease is observed after \sim 50 C. The half life values are in agreement with those given in (5,7,11,12) obtained in accelerators with oil diffusion pumps, but they are lower than the results from vacuum systems with ionic pumps (7,12).

Deuterium titanium targets have given, generally, satisfactory performance. Typical results of the relative neutron yield versus target loading are shown in Figure 6 for different operating conditions of the accelerator; it is evident from this figure that about 70 coulombs (\sim 12 C/cm²) are required to reach the saturation with an average current of 200 μ A (9 mA peak current) at 145 kV; each day, at the start of operation, the neutron yield increases slightly. The yield of the D-D reaction increases with time as a self-loading target, by the inclusion of deuterium in the titanium or, more probably, in the

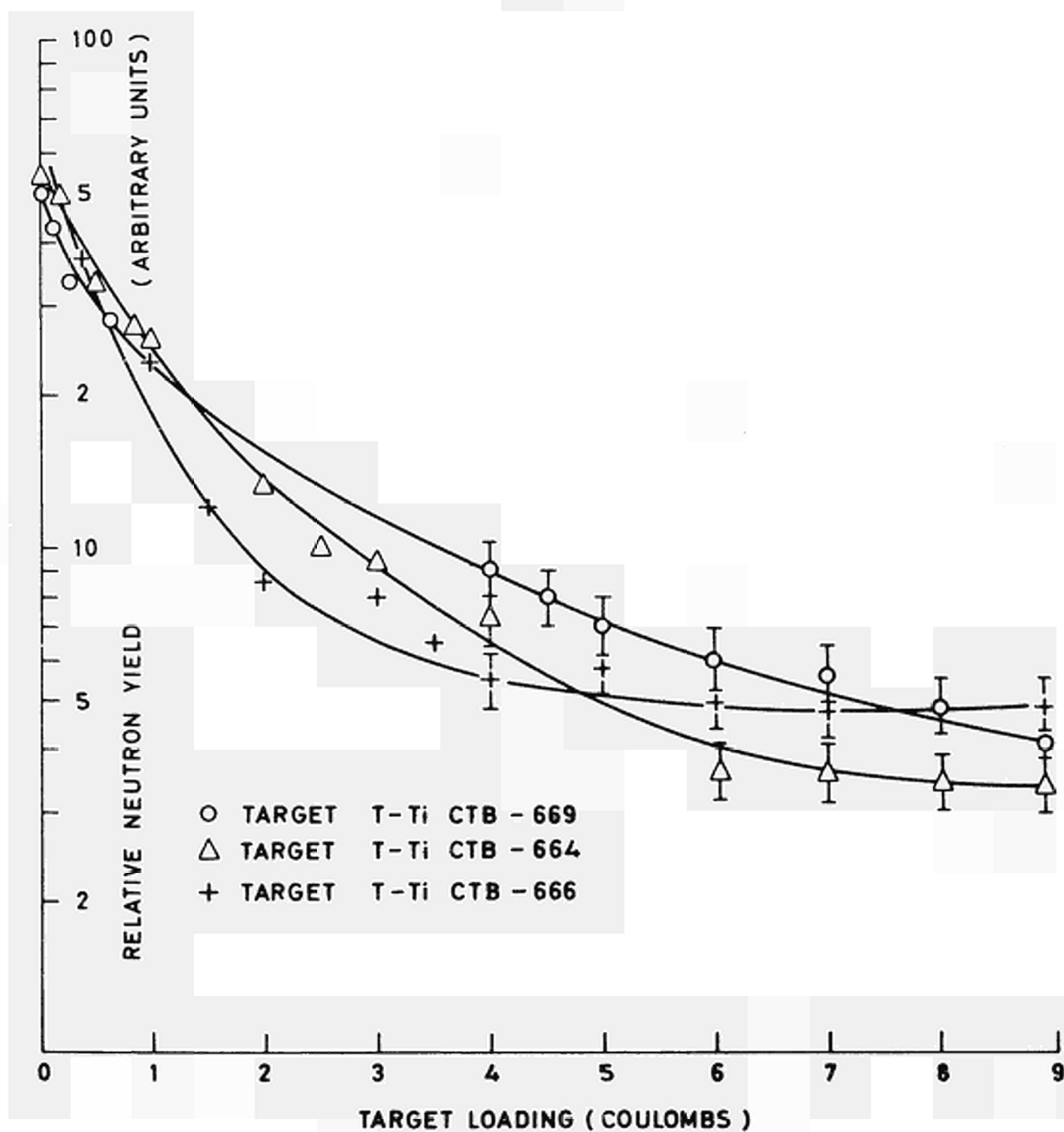
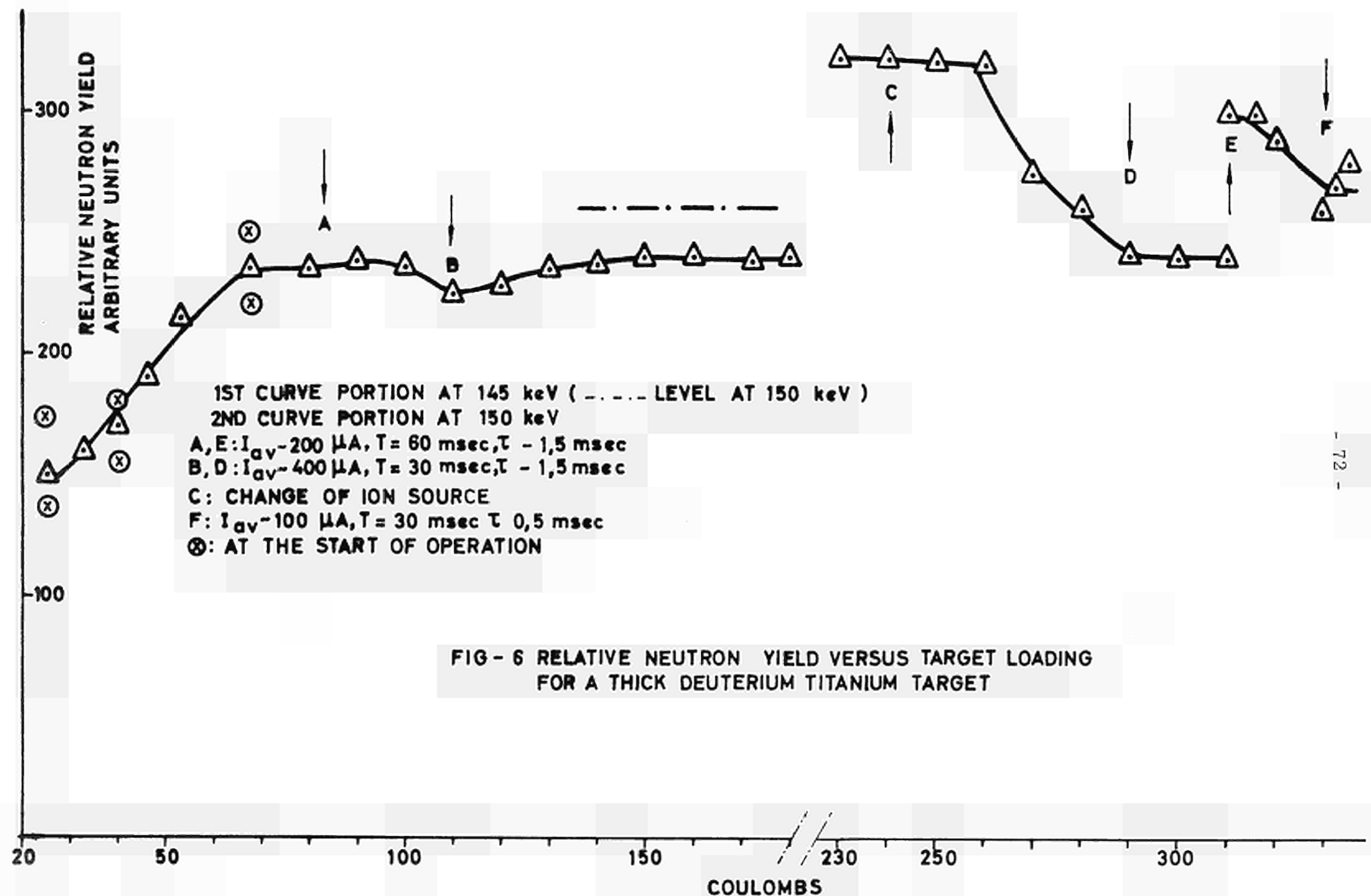


FIG. 5.- RELATIVE NEUTRON YIELD VERSUS TARGET LOADING FOR DIFFERENT TRITIUM TARGETS.



contamination layer, from deuterons impinged on the target. Afterwards the yield remains almost constant; we assume that a saturated thick target of carbon self-loaded with deuterium has been formed. In a first approximation the curve of figure 6 is usually independent of the beam current, however, there is a neutron yield fall of order of 10% when increasing the repetition rate by a factor of 2 (B and D portions), due, probably, to increasing surface temperature. The yield also falls about 10% when changing the pulse length from 1,5 to 0,5 msec., due perhaps, to the change in the average ratio of atomic to molecular ions and the focusing of the beam (see the pulse shape at 1st. portion, Figure 3B). The neutron yield varies about 20% with the state of the ion source (C portion); initially, when the ion source is clean it is possible to obtain higher atomic ion ratio. The measurements with this target were extended over a period of three months and they have been shown to remain constant within 20 per cent. From our results with the D-D reaction on carbonaceous multilayers, we think that this method will probably give a promising material for self-loading targets.

In view of the above measurements it is concluded that our vacuum system is appropriated for work with carbon multilayer self-loading targets; other vacuum pumps (ionic or turbomolecular pumps) are necessary, however, for using tritium-titanium targets.

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CIBLES A HAUT RENDEMENT NEUTRONIQUE POUR
UN ACCELERATEUR LINEAIRE A ELECTRONS
(FAISCEAU A PUISSANCE MOYENNE ELEVEE
DE L'ORDRE DE 10 kW)

C. ALLARD

Bureau Central de Mesures Nucléaires
EURATOM, Geel, Belgique

ABSTRACT

High Neutron Yield Targets for a Linear Electron Accelerator

A brief definition is first given of the concept of the neutron yield of a target and the difficulty underlined of using all the power of an electron beam for the production of neutrons with 100% yield.

The more the power increases, the more necessary it is to break up the heavy neutron-generating metal (uranium) in order to cool it, and the fraction of the cooling liquid becomes of ever greater relative importance, to the detriment of the neutrons produced.

A description is then given of the principle of the target used at the BCMN, in which a special uranium arrangement makes it possible to obtain the maximum amount of neutrons with a high-power electron beam (10 kW). A description is also provided of the auxiliary rigs essential for this type of target, together with their measuring and safety circuits.

Finally, the advantages and drawbacks of this type of target and the anticipated lines of future development are sketched on the basis of the latest results obtained.

I. Introduction

La cible du BCMN dont je vais vous parler a de commun avec les cibles qui ont été décrites précédemment le fait qu'elle est également destinée à produire des neutrons à partir d'un accélérateur.

Mais, comme il s'agit chez nous d'un accélérateur linéaire à électrons dont l'énergie moyenne est de l'ordre de 50 MeV, sa conception est toute différente de celles-là; il s'agit en effet d'une cible "épaisse" en uranium naturel dans laquelle on envoie le faisceau pulsé d'électrons du Linac qui s'y trouve entièrement absorbé. Les électrons "freinés" dans l'uranium émettent un rayonnement γ (c'est le Bremsstrahlung) qui produit des neutrons par deux types de réactions:

pour les 2/3 par réaction $(\gamma, N) = \text{photoneutrons}$

pour 1/3 environ par réaction $(\gamma, f) = \text{neutrons de fission}$

(4). Ceci permet de supposer que les cibles utilisées avec un accélérateur linéaire posent des problèmes qui leur sont très particuliers et que nous allons examiner.

II. Problèmes posés

La conception de la cible est étroitement liée à son utilisation et donc aux types de mesures neutroniques qui sont faites à partir d'elle.

C'est aussi ce type de mesures qui a fixé les caractéristiques de l'accélérateur auxquelles elle doit être adaptée.

Dans le cas du BCMN, l'accélérateur délivre actuellement un faisceau d'une puissance moyenne garantie de 4,2 kW. Cette puissance doit par la suite atteindre 10 - 12 kW.

Notre cible devait donc être conçue pour d'abord absorber sans dommage une puissance voisine de 4 kW, puis ensuite sa structure devait évoluer vers une cible capable d'absorber toute la puissance disponible du faisceau.

Il y a donc deux problèmes:

- l'un lié au type de mesures,
- l'autre lié à la puissance du faisceau.

- Adaptation au type de mesures:

Les mesures faites au Linac du BCMN (1) étant essentiellement des mesures neutroniques par la méthode du temps de vol (mesures de sections efficaces totales, de capture, de fission etc...), la cible devait être conçue (5) de façon à ce qu'elle puisse être, du point de vue neutronique, assimilée approximativement à un point, donc, avoir des dimensions réduites. Par ailleurs sa structure ne devait pas être la cause, par diffusion élastique des neutrons à l'intérieur, d'un temps de relaxation élevé de ceux-ci.

Comme elle devait être entourée d'un ralentisseur dans la plus part des cas elle devait donc être assez dégagée dans la salle des cibles de toute masse métallique.

Les mesures sont en effet effectuées selon des directions privilégiées (bases de vol) qui viennent converger vers le centre de la cible.

Leur précision dépend notamment de la précision avec laquelle on mesure le temps de parcours des neutrons entre le centre d'émission des neutrons l'échantillon placé à une distance donnée le long de la base de vol correspondante et les détecteurs associés.

- Adaptation de la cible à l'accélérateur:

- Sans entrer dans les détails il est bon, je pense, de parler très rapidement des caractéristiques de notre accélérateur pour pouvoir comprendre les exigences auxquelles la cible doit s'adapter.

Les caractéristiques de fonctionnement de notre accélérateur tant en impulsions courtes (1000 Hz - 10 ns à 50 ns) que longues (250 Hz - 2 μ s) conduisent actuellement à un faisceau de puissance de l'ordre de 4 kW ceci

pour un fonctionnement des klystrons à 12 MW - 8 kW moyens. Le constructeur de l'accélérateur s'est engagé à porter ces dernières valeurs à 24 MW - 16 kW ce qui devrait nous donner un faisceau de 10 - 12 kW moyens.

Nous devons donc bien sûr faire évoluer notre cible pour qu'elle puisse accepter les puissances correspondantes.

- Dans tous les cas le faisceau a un diamètre qui n'excède pas 1 cm et théoriquement 80% de la puissance du faisceau se trouve située dans un diamètre de 0,5 - 0,6 cm, ces dimensions peuvent être même plus faibles dans le cas d'impulsions de 1 à 2 μ s. Ceci permet de fixer le diamètre de la partie active de la cible dans laquelle le faisceau est ralenti; il doit pouvoir admettre un décentrage de celui-ci de ± 1 cm, ce qui est réalisé avec un diamètre de 2,5 à 3 cm.
- Il existe une disposition particulière de l'optique de la sortie de faisceau de l'accélérateur (1) et (2) qui permet, en plus du tir horizontal classique, un tir vertical sur une cible dont l'axe doit être également vertical. Cette dernière disposition a été adoptée afin de diriger vers le sol le γ -flash qui, dans certains cas, perturbe la détection des neutrons et la qualité des mesures. Par ailleurs on arrive ainsi à une parfaite symétrie neutronique des bases de vol qui sont alors toutes équivalentes.

III. Solution adoptée au BCMN

Il a déjà été décrit le principe général de l'installation de la cible du BCMN dans le Proceeding de la première Conférence (2) (1964) aussi vais je me borner à en rappeler les points essentiels.

1. La nécessité d'avoir deux directions de faisceau (horizontale et verticale) nous avait fait d'abord envisager l'utilisation d'une cible orientable. Ceci a été abandonné pour divers raisons technologiques. Nous avons de préférence retenu la solution de deux cibles séparées comme il est montré sur la figure 1. Ces deux cibles doivent nécessairement être interchangeables et sont placées, au choix, sur un ascenseur au sommet duquel elles viennent indifféremment s'embrocher. Ce principe a impliqué la réalisation d'une cellule de stockage de cibles (4 au maximum) alimentée par un chariot télécommandé (voir fig. 2 et photos 2a et 2b.)

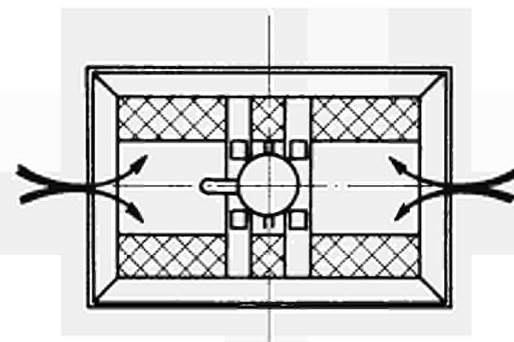
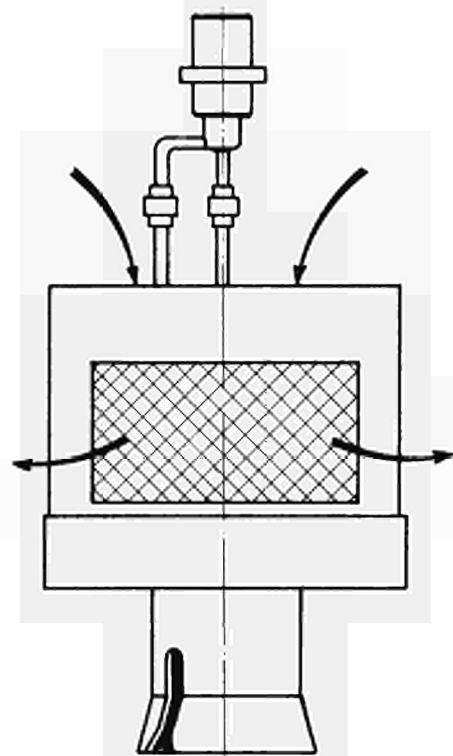
2. En 1964, j'avais décrit un type de cible (2), dérivé de celle utilisée à Saclay, qui devait alors être essayée sous peu au BCMN.

Depuis cette date on peut dire que cette cible a donné satisfaction et que la puissance moyenne maximum qu'elle a acceptée sans dommage a été de l'ordre de 4 kW.

Après 2500 H d'utilisation à des puissances inférieures à 4 kW, on a retrouvé la fenêtre en contact avec l'uranium déchirée à la suite vraisemblablement d'un décentrage prolongé du faisceau. Aucune contamination immédiate n'a été décelée et, grâce à notre dispositif de manutention et de stockage, la cible a été immédiatement remplacée dans l'heure sans qu'il s'en suive d'autre inconvénient.

3. Je crois utile cependant de faire un retour en arrière pour permettre de comprendre l'évolution que nous avons suivie et le but que nous nous proposons d'atteindre.

N'ayant pas eu la possibilité de faire d'études systématiques de cibles et afin de pouvoir utiliser assez rapidement notre accélérateur, nous sommes partis d'une cible de conception simple qui avait fait ses preuves à Saclay à une puissance maximum de faisceau voisine de 2 kW.



VERTICAL TARGET UNIT

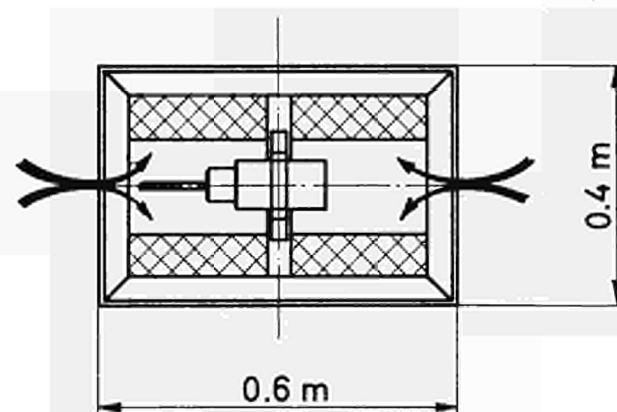
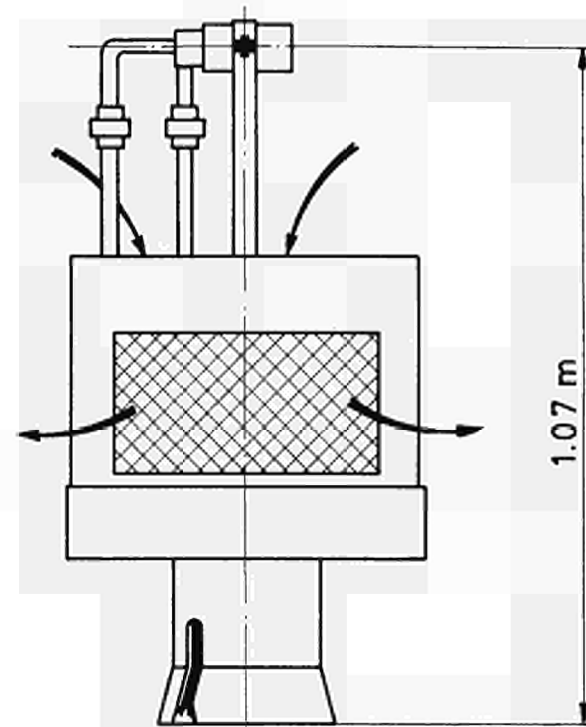
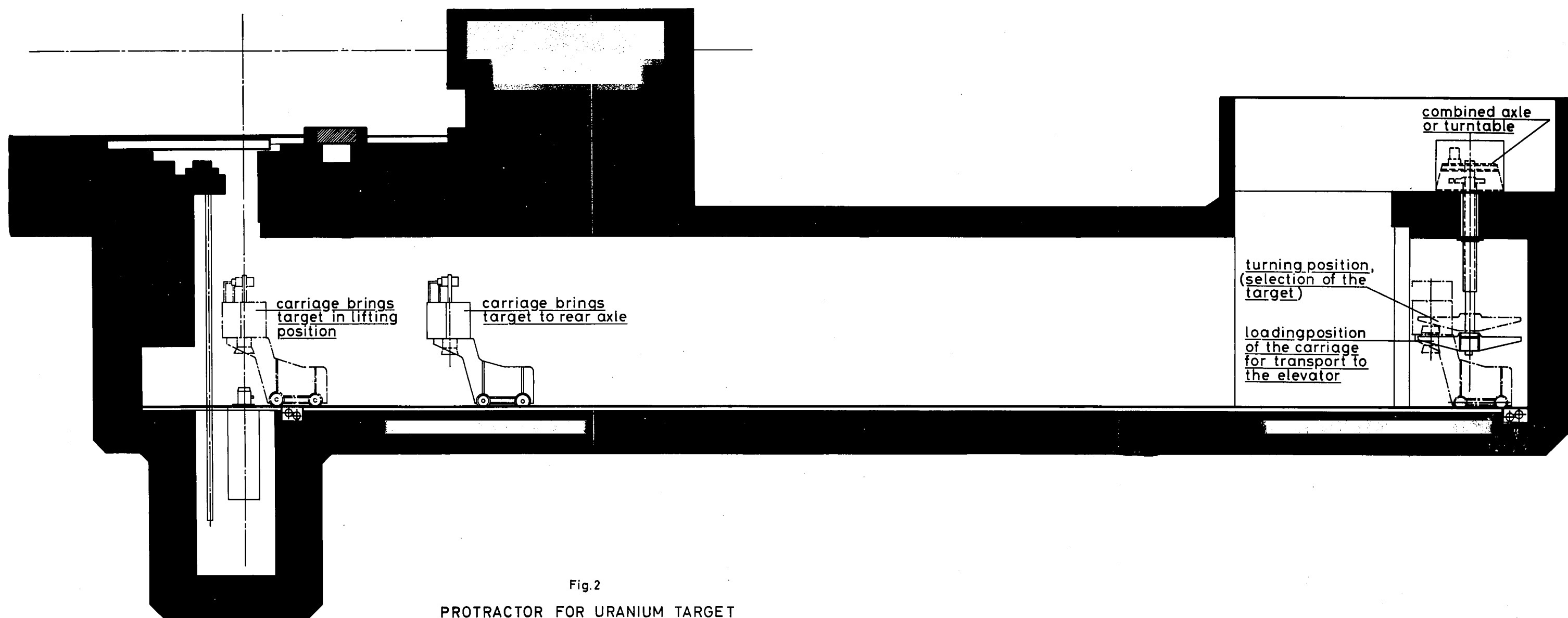


Fig.1

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HORIZONTAL TARGET UNIT



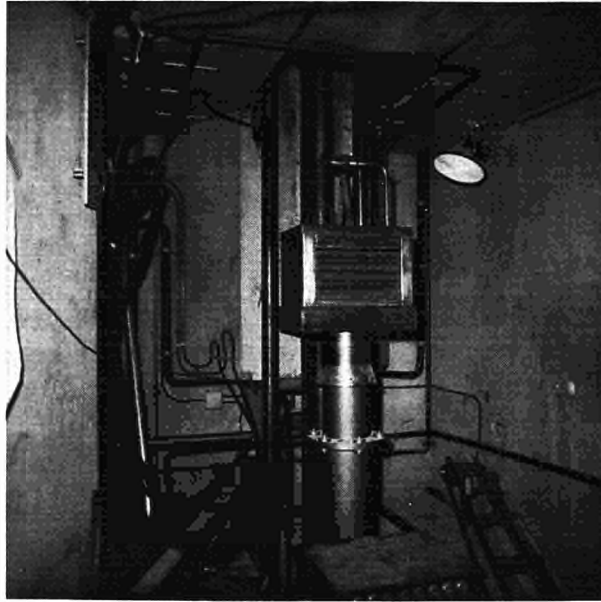


FIGURE 2a

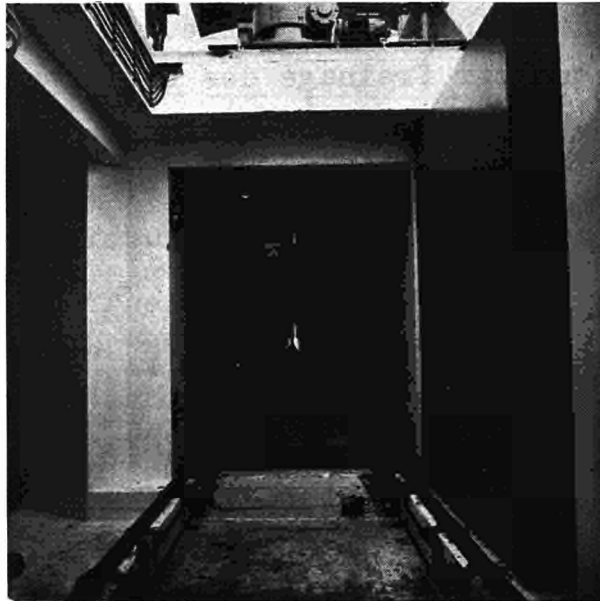


FIGURE 2b

Comme dans la cible de Saclay (fig. n° 3-I), nous avons utilisé un noyau cylindrique d'uranium de $\varnothing = 3$ cm et de 10 cm de long. Nous avons conservé l'étalement du faisceau (4) entrant dans l'uranium, par contact sur une surface conique qui évite un impact trop direct, donc diffuse le point chaud.

Nous allons voir qu'une telle cible peut être considérée comme possédant une épaisseur infinie donc d'un rendement neutronique théorique maximum pour une puissance de faisceau compatible avec des conditions de refroidissement qui permettent à l'uranium de rester en phase stable.

- Résultats connus:

Il n'est pas dans mon intention de faire un exposé théorique sur les cibles mais simplement de rappeler un certain nombre de résultats généralement admis (3) qui permettent de définir le rendement neutronique d'une cible. On sait en effet que le nombre de neutrons par freinage des électrons dans la matière est fonction:

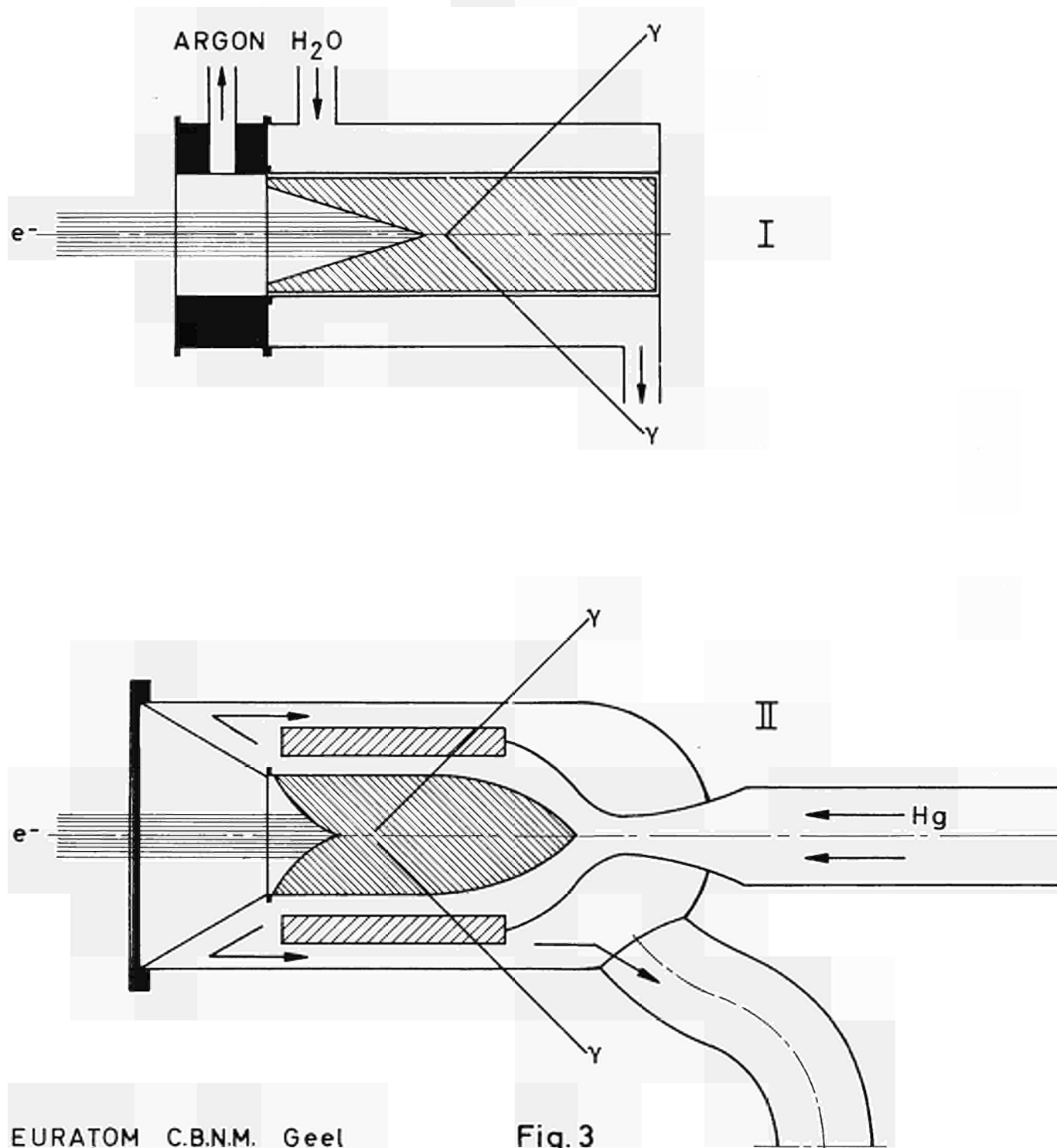
- $N=f(Z)$: du choix de cette matière, les noyaux lourds permettant la plus grande production de photoneutrons.

- $N=f(\lambda)$: Par ailleurs ce nombre de neutrons est fonction de l'épaisseur de la matière se trouvant sur le chemin du faisceau d'électrons et que ce nombre est pour une puissance de faisceau donnée, pratiquement constant au delà de 10 longueurs (λ) de radiation.

On peut dire qu'au delà de 10 λ la cible est équivalente à une cible d'épaisseur infinie.

Si l'on prend comme référence l'uranium naturel, dans les conditions d'une cible d'épaisseur infinie le flux de neutrons produit (par réactions γN , γf) est de:

$$2,6 \cdot 10^{12} \cdot N \cdot \text{sec}^{-1} \cdot 4\pi \cdot \text{kW}$$



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Fig. 3

une telle cible, capable d'accepter sans détérioration un faisceau de 1 kW aura d'après notre définition un rendement neutronique de 100%.

- Nous avons vérifié expérimentalement que le rendement de cibles en Pb et en Hg "d'épaisseur infinie" avaient des rendements voisins de 50%. Nous pensons qu'il devrait en être de même pour le W.
- Toujours dans les mêmes conditions il est admis que le rendement neutronique d'une cible en uranium enrichi (^{235}U à 93%) aurait un rendement de 160%.

- Evolution suivie:

Notre but était donc de conserver le rendement de 100%, que possède, par sa conception, la cible n° I (fig. 3) à une cible capable d'absorber un faisceau jusqu'à 10 kW; ceci toujours avec le souci d'utiliser à 100% le faisceau pour la production des neutrons.

C'est pourquoi nous avons opté au départ (5) pour la construction d'une cible dont le noyau d'uranium naturel avait les mêmes dimensions mais dont nous avons essayé d'améliorer les conditions de refroidissement.

Comme nous avons par ailleurs, dans l'avenir, l'intention de passer à un noyau en uranium enrichi à 93%, nous avons dès le départ éliminé l'eau comme fluide de refroidissement qui possède pour les neutrons un pouvoir réflecteur dangereux.

Nous lui avons préféré le mercure, dans lequel d'ailleurs comme nous l'avons dit, le faisceau peut, en se ralentissant, produire une quantité appréciable de neutrons.

C'est pour ne pas faire baisser le rendement de la cible que nous n'avons pas retenu un noyau en tungstène ou tantale qui auraient pour une fiabilité meilleure donné 50 à 60% moins de neutrons.

Nous n'avons pas voulu réaliser une cible où le noyau aurait été divisé pour mieux le refroidir car, dans ce cas, le fluide de refroidissement se substitue à l'uranium et le taux de neutrons diminue, ou alors pour le maintenir on est conduit à augmenter les dimensions de la cible.

Nous avons donc essayé d'arriver à la limite de l'utilisation d'un noyau homogène ce qui bien sûr augmente la possibilité d'incident.

- Incidents possibles:

La qualité de la cible dépend de sa tenue à un certain nombre d'incidents possibles:

1. Tenue thermique du noyau:

- Le noyau d'uranium ne doit pas fondre ce qui implique un bon contact thermique entre le fluide de refroidissement et l'uranium et un dimensionnement adapté à la puissance à évacuer.
- Le gainage ne doit pas se rompre: soit par dilation différentielle immédiate, soit à long terme, sous l'action des cycles thermiques par une évolution de la structure de l'uranium qui entraîne, selon un processus difficile à prédéterminer, un allongement progressif et irréversible des cotes du noyau.

2. Tenue thermique de la fenêtre:

Un faisceau de quelques kW dissipe près de 100 W dans une fenêtre en acier inoxydable de 5/10 de mm d'épaisseur.

La tenue de cette fenêtre dans le temps à la température et aux modifications de structure de l'acier sous l'action du bombardement électronique est un élément à la fois difficile à évaluer et vital pour la qualité de la cible.

Nous nous sommes donc efforcés de perfectionner la cible de départ n° I en la faisant évoluer vers les cibles:

- N° II: - meilleur refroidissement de la fenêtre,
(fig.3) - diminution de l'épaisseur du gainage de l'uranium ($e = \frac{5}{10}$ au lieu de 1 mm),
- contact thermique entre l'U et sa gaine réalisé par l'He à 300 g/cm² absolus,
- stabilisation de l'uranium par 10% de Mo,
- pose d'un thermocouple pour mesurer la température de l'uranium près du point chaud du noyau,

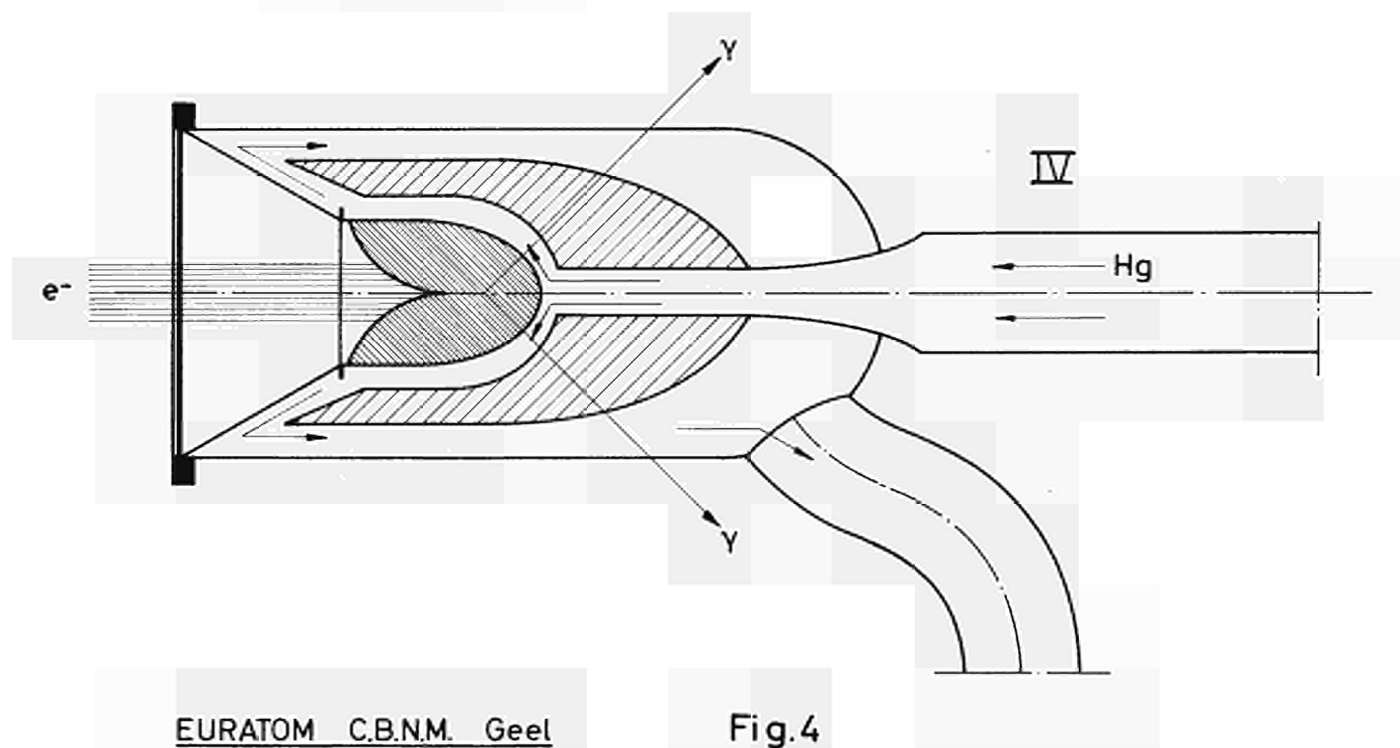
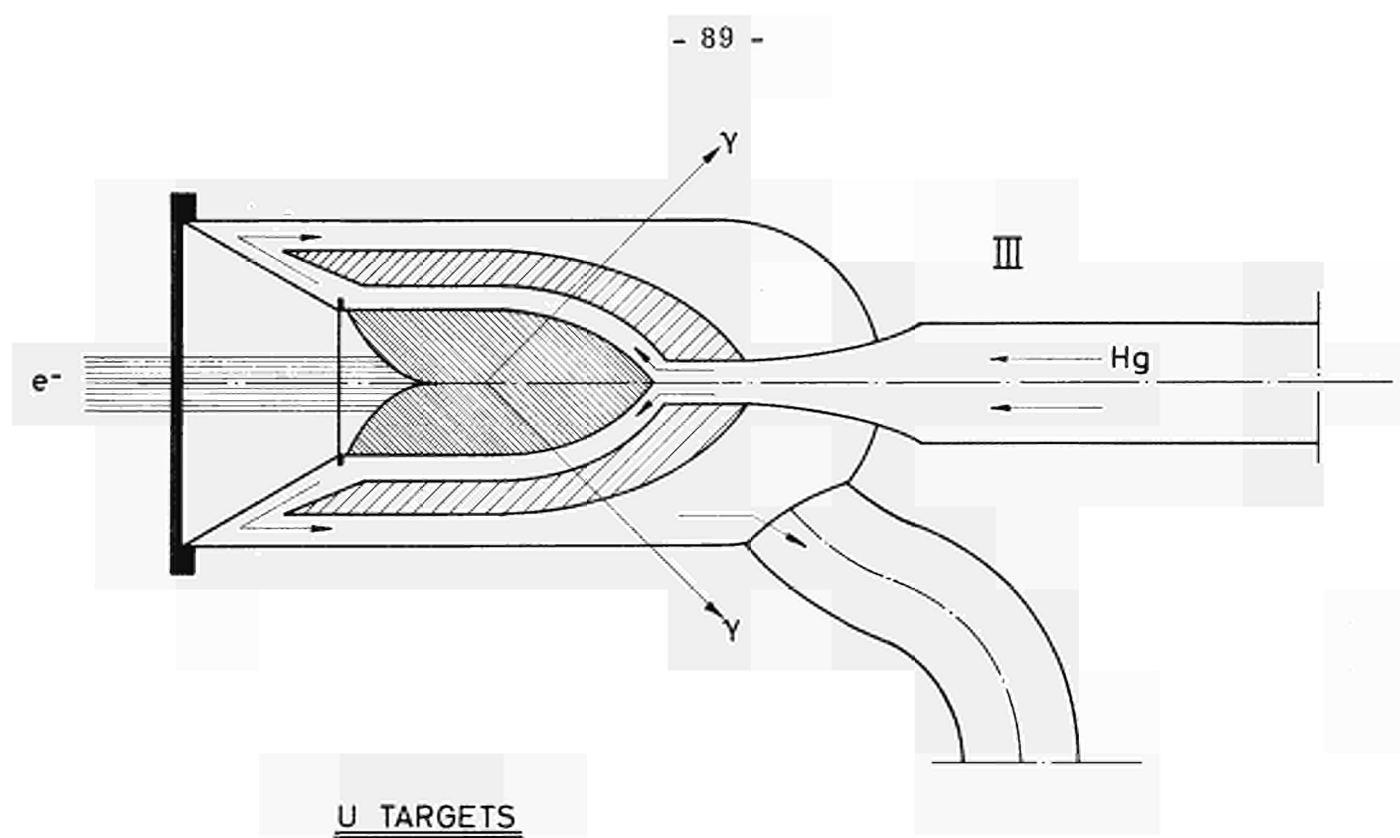
- meilleure absorption des γ grâce à une couronne cylindrique d'uranium entourant le noyau (5).

N° III: - On conçoit intuitivement que pour un débit de mercure donné par la pompe existante, il existe (fig.4) une puissance maximum au delà de laquelle le noyau d'uranium, dans lequel la puissance du faisceau est absorbée, va fondre. C'est cette puissance maximum qui fixe les limites d'utilisation de la cible.

- Tout le problème consiste à donner à ce noyau des dimensions telles que le fluide de refroidissement se rapproche du point chaud théorique (ici estimé à 6 mm en arrière de la pointe du cône d'entrée du faisceau) tout en conservant au noyau une surface de refroidissement compatible avec le nombre de W/cm^2 qui peuvent être évacués par le mercure.
- On pourrait être tenté par une réalisation du type IV (fig. 4) où le noyau a juste 6 longueurs de radiation et où le mercure encercle totalement la zone chaude de l'uranium. On pourrait croire que l'on est ainsi dans les meilleures conditions pour son refroidissement.

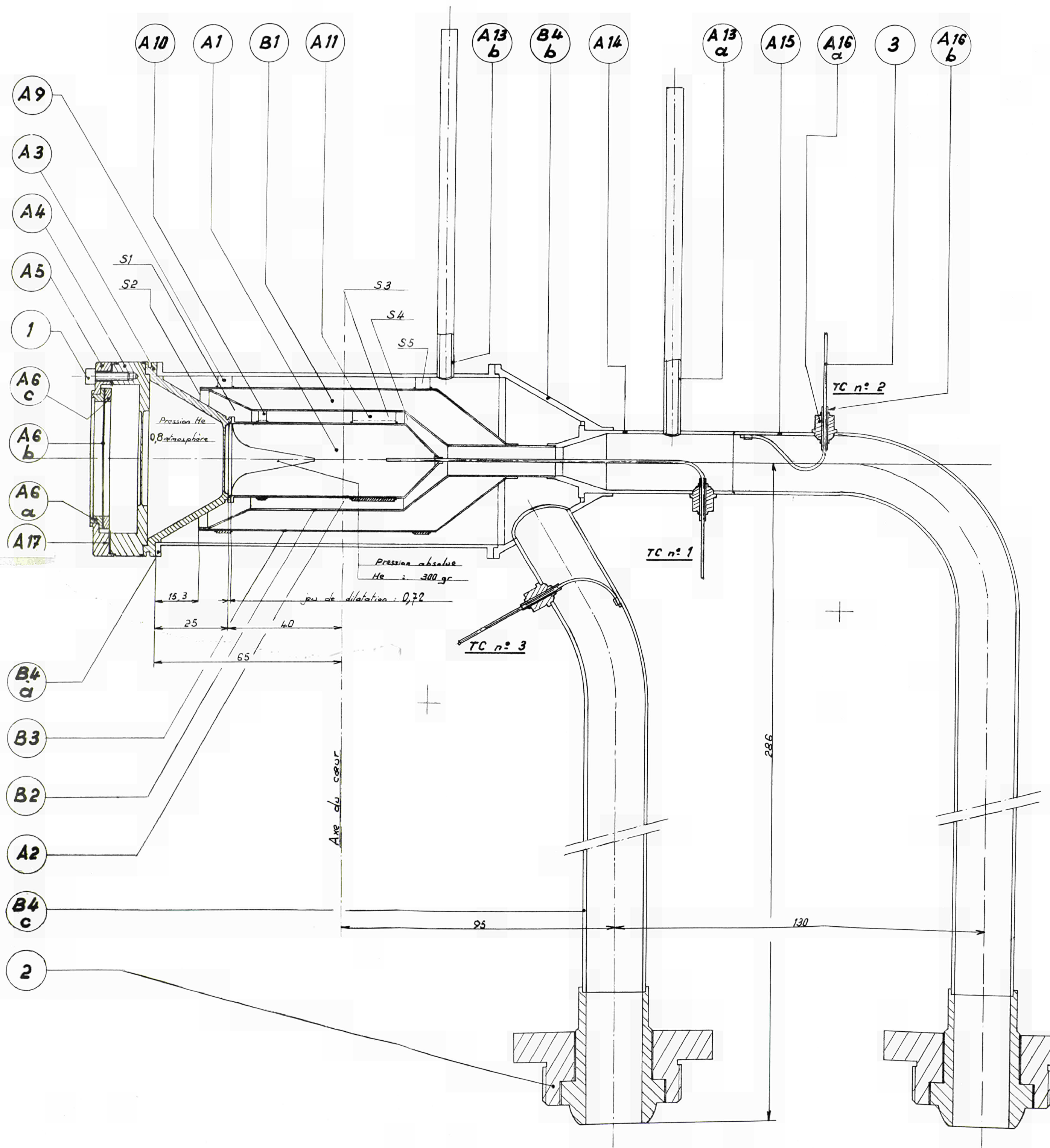
Nous n'avons pas opté pour cette solution extrême car elle réduisait la surface de refroidissement du noyau à quelques dizaines de cm^2 ce qui aurait impliqué que l'on puisse évacuer, à 10 kW, plus de $500 W/cm^2$ de surface en contact avec le mercure. A notre connaissance cela semble très improbable.

- Aussi avons nous (6) réalisé (fig. 5 et photos 5a et 5b) le noyau en retenant le type n° III où l'on se situe dans des limites plus classiques:
 - Diamètre du noyau: 2,5 cm.
 - Profondeur de pénétration maximum possible pour le faisceau: 4 cm.
 - Surface de refroidissement: $55 cm^2$



EURATOM C.B.N.M. Geel

Fig.4



Sections de passage Hg

S1	: 445 mm ²
S2	: 425 mm ²
S3	: 260 mm ²
S4	: 240 mm ²
S5	: 450 mm ²

3	Thermo couple	3	Réference	PABAC 10 T1 long. 2 m
2	Raccord Eucatom	2	Inox	existant
1	Vis CHc M3 - 12	3	Inox	
A17	Joint ϕ 52 x 66 ep. 0,3	1	Cuivre	
B4c	Tube sortie Hg	1	Z 3 CN 18/10	
B4b	Fond conique	1	"	
B4a	Enveloppe	1	"	
B3	Gaine ext. tube	1	"	
B2	Gaine inter. tube	1	Z 3 CN 18/10	
B1	Tube	1	U Mo	
A16b	Canon + minitube 2 x 0,5 lq. 15	3 x 3	Z 3 CN 18/10	
A15	Tube d'arrivée Hg	1	"	
A14	Cône d'entrée Hg	1	"	
A13b	Tube de pompage aval	1	"	
A13a	Tube de pompage amont	1	"	
A11	Entretoise AR petit ϕ	1	"	
A10	Entretoise AV petit ϕ	1	"	
A9	Entretoise grand ϕ	2	Z 3 CN 18/10	
A6c	Rondelle AR	1	Z 3 CN 18/10	
A6b	Membrane ϕ 49 ep. 0,2	1	Z8 CNNb 25/20	
A6a	Rondelle AV	1	Z3CN 18/10	
A5	Porte membrane amovible	1	"	
A4	Support AV	1	Z 3 CN 18/10	
A3	Porte membrane conique	1	Z8 CNNb 25/20	
A2	Gaine du cœur	1	Z 3 CN 18/10	
A1	Cœur	1	U Mo 10 %	
Rep.	Désignation	Qté	Matière	Observations

REPRODUCTION ET COMMUNICATION INTERDITES SANS NOTRE AUTORISATION (LOI DU 11 MARS 1902)

COMPAGNIE POUR L'**E**TUDE ET LA **R**ÉALISATION DE **C**OMBUSTIBLES **A**ТОMIQUES

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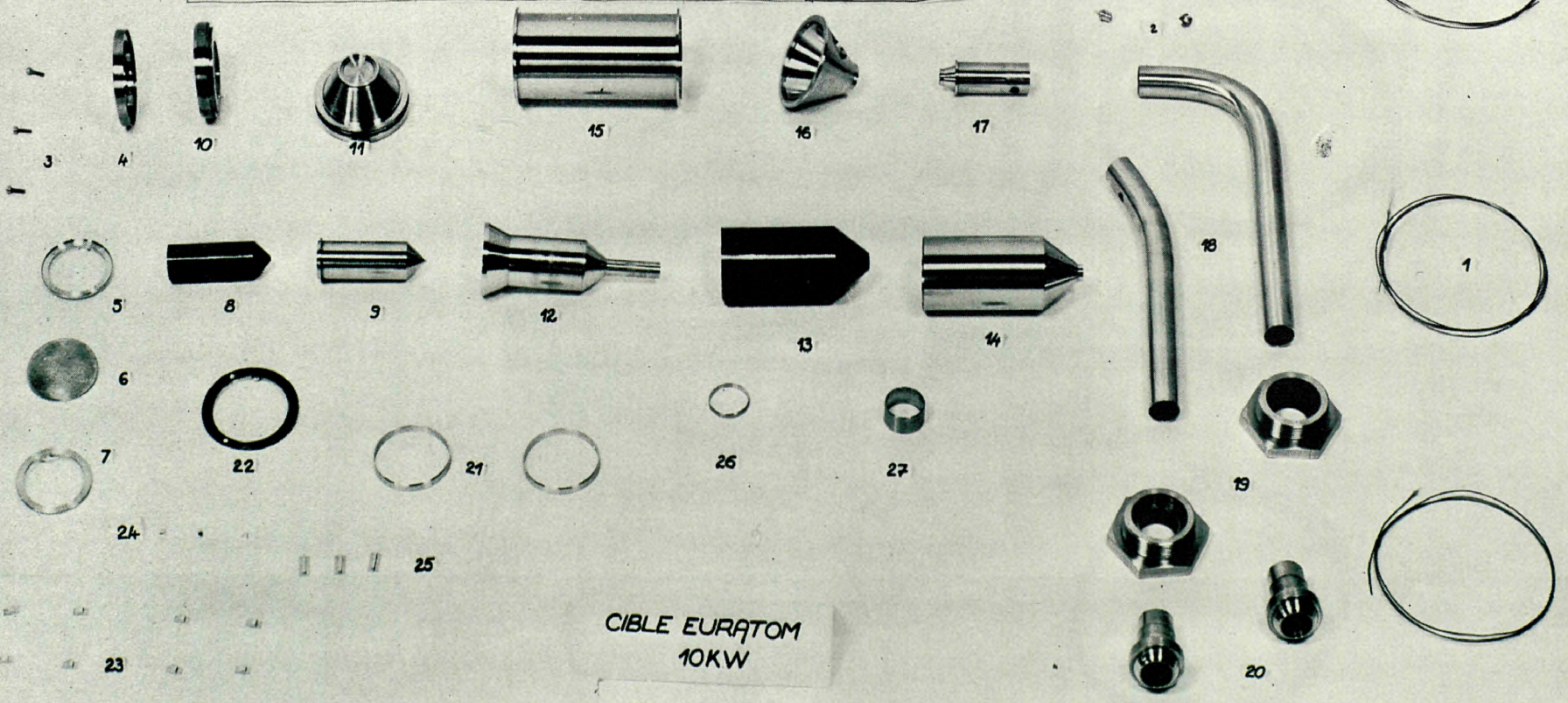
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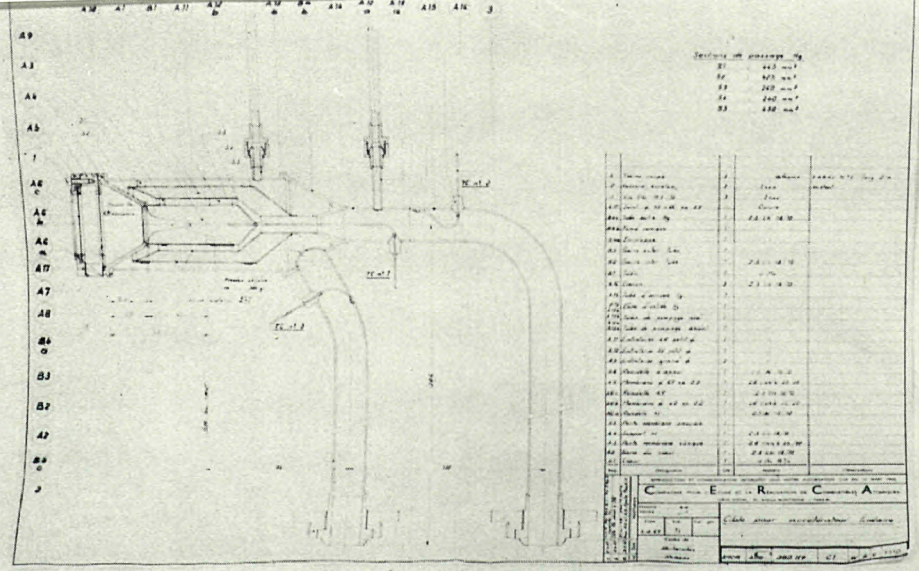
Cible pour accélérateur linéaire

Fig. 5



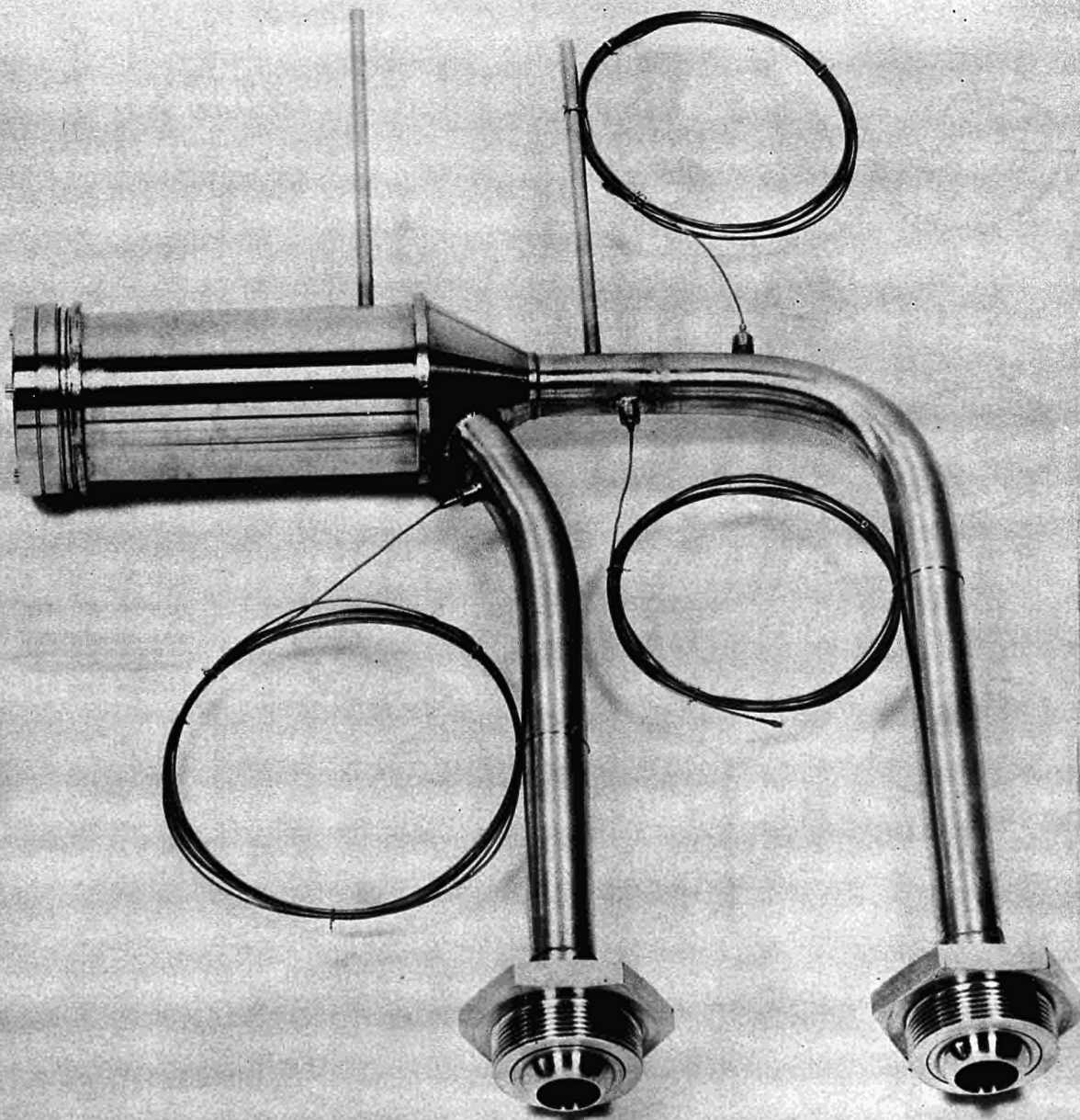
CIBLE EURATOM
10KW

FIGURE 5a



CIBLE EURATOM
10KW

FIGURE 5b



ce qui fait à 10 kW : 180 W/cm^2

et une température mesurée de 305° (relevée à 5 kW à 1,4 cm derrière le point chaud).

Compte tenu du coefficient de conductibilité thermique de l'U + 10% à $T > 600^\circ$ que l'on donne pour:

$$\pm 400 \text{ W/cm}^2/\text{cm}/^\circ\text{C}/\text{sec}$$

on arrive ainsi à une température théorique du point chaud de 650° à 5 kW et par extrapolation:

950° à 8 kW

1200° à 10 kW.

On voit que dans l'état actuel des choses, à 10 kW, on serait très près du point de fusion de l' U^{238} + 10% Mo qui est de 1250° et c'est ce qui fait que nous ne pensons pas prudent de dépasser 7 - 8 kW avec la cible actuelle.

Cependant, grâce à une modification possible du circuit de refroidissement nous comptons, assez facilement, atteindre les 10 kW.

Il faut noter que, dans notre réalisation, le noyau est pratiquement entièrement entouré d'une coupole d'uranium qui capte la presque totalité des γ émis dans le noyau. Ce dispositif permet de bénéficier au maximum des réactions (γ, N) (γ, f) pour la production de neutrons et réduit considérablement le γ -flash dans l'axe du faisceau.

- La fenêtre double a été choisie en acier inox stabilisé (Z8 CNN6 25/20) et usinée dans la masse pour éviter les soudures. Comme on le voit sur le plan d'ensemble (fig.5) l'espace interfenêtres est rempli d'He à 800 g absolus afin d'assurer un bon transport de la chaleur dissipée dans la fenêtre et maintenir une atmosphère neutre au moins des deux côtés de la fenêtre de 5/10 de mm qui protège le noyau d'uranium.
- Nous n'avons pas retenu une solution où la fenêtre serait directement refroidie par le Hg car le risque est trop grand, en cas de rupture de cette fenêtre, de voir le mercure projeté dans la salle des cibles, la contaminant ainsi pour un temps indéterminé.

Par ailleurs cette solution serait la cause d'une baisse de rendement neutronique.

- Mesures et sécurités installées:

- La cible est à une soixantaine de mètres du pupitre de contrôle du fonctionnement de l'accélérateur. Elle est tout d'abord observée, en vue indirecte, par caméra de télévision.
- Un système de sécurité coupe le faisceau de l'accélérateur: en cas d'absence de ventilation de l'échangeur, en cas d'arrêt du débit de pompe électromagnétique, en cas de fuite de mercure.
- Du pupitre de contrôle on peut:
 - évaluer le débit de la pompe (dont on lit le courant d'alimentation qui est de l'ordre de 2000 A),
 - lire sur un voltmètre digital la température du noyau de l'uranium et suivre son évolution en fonction soit de la puissance du faisceau soit, indication intéressante, de son centrage,
 - la température d'entrée et de sortie du mercure dans la cible.

- Récupération d'une cible détériorée:

Afin de pouvoir récupérer l'ensemble échangeur-pompe électromagnétique, en cas d'incident sur la cible proprement dite, celle-ci se trouve connectée aux tuyaux de refroidissement de l'échangeur par l'intermédiaire de raccords spéciaux étanches au mercure (jusqu'à 150°C) (voir photos 5a et 5b).

Grâce à ce système on peut remplacer l'élément détérioré par un élément identique après un certain nombre d'opérations: vidange de la cible détériorée, remplissage de mercure de la nouvelle.

Ces opérations se font évidemment après que l'on ait laissé désactiver l'ensemble un temps suffisamment long et nécessitent beaucoup de précautions.

IV. Conclusions

L'option que nous avons choisie au départ nous a amené à un type de cible dans lequel le rendement neutronique est maximum. Nous avons le sentiment que la solution choisie a des limites qui ne permettront pas de dépasser 10 kW de faisceau, limite probable des possibilités immédiates de notre accélérateur. Elle nous convient donc actuellement.

Quand nous aurons, après une longue utilisation de cette cible, acquis la certitude de son bon comportement sous faisceau, nous remplacerons le noyau actuel par un noyau en U^{235} . Nous croyons ainsi obtenir avec de faisceau 10 kW un flux de:

$$\phi = \pm 4.10^{13}.N.S^{-1}.4\pi.$$

Si par la suite nous voulons doubler les caractéristiques de notre Linac il nous faudra adopter une autre solution qui obligatoirement nous obligera à diviser le noyau d'uranium pour mieux le refroidir ce qui de ce fait diminuera le rendement neutronique de la cible, à moins qu'une solution, actuellement à l'état de projet, permette d'atteindre les 20 kW sans changer la structure de la cible.

Il s'agirait d'une cible refroidie à l'He gaz à haute pression (10 atm) à grand débit, dans laquelle l'uranium serait gainé d'Alu comme un élément de pile. Les premiers calculs semblent montrer qu'une telle solution n'est pas une utopie. Sa description pourrait peut-être faire l'objet d'une communication lors de la 4^{ème} Conférence sur "les cibles pour accélérateurs destinées à la production de neutrons".

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G.C.Baldwin, E.R.Gaerttner, M.L.Yealth, Phys.Rev. 104, 1652, (1956).
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- (5) K.H.Böckhoff, BCMN, communication privée.
- (6) Avec la collaboration de J.M.Salomé, BCMN.

Remarques: 1. Les fig. 1 - 2 et photos 2a et 2b reproduisent l'échangeur et le système de manutention de cibles conçus et réalisés en collaboration avec les firmes CCM - CSF et ERAC.

2. Les fig. 5 et photos 5a et 5b ont été aimablement communiquées par la Société CERCA qui a construit la cible.
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DISCUSSION

Mr. SMITH

I would like to confirm that you are using natural Uranium targets.

Mr. ALLARD

Nous utilisons en effet actuellement une cible en Uranium naturel mais je profite de l'occasion pour vous dire que notre intention est, dans l'avenir, de remplacer l'uranium naturel par de l'uranium enrichi (à 93%) quand nous serons certains de la tenue de la cible. Cela nous fera gagner un facteur 1,6 dans la production des neutrons.

Mr. SMITH

I presume you are aware of the system in use at Harwell, England.

Mr. ALLARD

Nous connaissons le buster d'Harwell, ensemble sous critique qui possède un facteur de multiplication de 5 ou 6 . Nous n'avons pas retenu une telle solution car :

- 1) elle manquait de souplesse : l'installation est en effet très imposante et n'était pas adaptée à nos possibilités de tir horizontal et vertical -
- 2) Nous n'avions pas alors l'infrastructure technique suffisante pour concevoir et exploiter une cible sous critique qui nous faisait un peu peur.
- 3) Cette cible n'était pas adaptée aux mesures de temps de vol que nous comptons faire avec des impulsions de quelques nanosecondes car elle avait " un temps de relaxation " trop élevé d'au moins 80 nanosecondes , temps dû à la diffusion élastique des neutrons à l'intérieur même de cette cible.

Mr. EGGERMANN

Dans le projet de cible de puissance élevée vous avez parlé de deux possibilités. Vous nous avez exposé la 1ère. En quoi consiste la seconde ?

Mr. ALLARD

L'examen de la diapositive n° 7 montre en effet deux représentations du noyau de la cible dans lequel la puissance du faisceau est absorbée.

Dans la vue supérieure qui correspond à la cible réalisée et testée, le noyau a une profondeur égale à dix longueurs de radiation de l'uranium naturel. Ce noyau a été déterminé pour que la puissance évacuée par cm^2 de surface d'inox en contact avec le fluide de refroidissement n'excède pas 200 W/cm^2 , qui est une valeur limite d'après les spécialistes.

La vue inférieure représente une solution plus séduisante où le fluide de refroidissement arrive très près du point chaud théorique de cette cible. Cependant pour que cette solution soit valable il faudrait admettre que l'on puisse évacuer 400 à 500 W/cm^2 , ceci est peut-être possible mais nous n'avons pas momentanément voulu prendre le risque de réaliser une telle cible car, en cas d'impossibilité, le noyau aurait fondu, entraînant des conséquences imprévisibles.

Il ne faut pas oublier en effet que nous ne pouvons nous permettre de faire des essais destructibles de cibles qui donneraient d'avantage d'enseignements.

Mr. ALBERT

Pouvez-vous préciser quel est le flux de neutrons en $\text{n/cm}^2/\text{sec}$. derrière la cible en U.

Quelle est la répartition en intensité de ce flux de neutrons au voisinage de l'axe du faisceau juste derrière la cible en U dans des positions où il est possible de disposer un échantillon à irradier ?

Mr. ALLARD

Le flux de neutrons/ cm^2/sec , derrière la cible en U dépend de la distance au centre de la cible.

Le flux dans 4 m étant de l'ordre de $2,6 \cdot 10^{12} \text{ n/4 m/sec/kW}$ on peut faire aisément le calcul par cm^2 en sachant que :

- d'après des mesures absolues faites par M. NACHTIGALL, chef du service de Radioprotection du BCMN et MM. DUFFAR et VIALETTES du SPR du CEA, derrière la cible, le flux de neutrons est 0.7 fois celui que l'on trouve dans une direction perpendiculaire au faisceau **donc à l'axe de la cible**.
- Toujours derrière la cible (où le flash est minimum) on peut disposer des échantillons à une distance de 20 à 30 cm du centre neutronique de la cible.

- Par contre dans une direction perpendiculaire à l'axe de la cible (ou au faisceau) là où le flux de neutrons est maximum rien n'empêche de disposer des échantillons à quelques centimètres de la cible (5 cm)

Mr. LAVERLOCHERE

A quelle distance minimum de l'axe de la cible peut-on placer un petit échantillon ?

Mr. ALLARD

Cette distance est limitée par les dimensions mêmes de la cible qui a 5 cm de diamètre.

On peut mettre des échantillons tout près de la paroi de la cible si les physiciens n'ont pas placé de modérateur contre cette paroi.

La distance de 4 à 5 cm, pour un échantillon de quelques centimètres, est raisonnable.

Mr. BRUNINGHAUS

Können Sie etwas sagen über das Energiespektrum der Neutronen, welche Sie mit dem Urantarget erhalten ?

Mr. ALLARD

Le spectre d'énergie des neutrons produits par notre cible se compose :

- 1) du spectre de fission de l'uranium qui est connu et invariable (70 à 80% des neutrons produits doivent avoir une énergie comprise entre 800 keV et 1 ou 2 MeV.)
- 2) A ce spectre vient se superposer le spectre des photoneutrons qui s'étale de 2 MeV à 10 ou 15 MeV.

Il dépend de l'énergie du faisceau d'électrons incidents ralentis dans la cible. La queue de ce dernier spectre est assez indéterminée.

Mr. BOCKHOFF ici présent, responsable des mesures neutroniques faites autour de notre accélérateur sera mieux à même que moi-même de vous répondre sur ce point.

Mr. BOCKHOFF

Wir haben das Spektrum mit der time-of-flight-Technik und einem Plastik-Scintillator gemessen. Aus der yield Kurve ergibt sich ein Neutronenspektrum

gleich einem Verdampfungsspektrum mit einem Maximum zwischen 1 und 2 MeV. Das Maximum ist nicht genau das Ende des Spektrums, dieses ist nicht feststellbar, weil der Detektor durch den Gammaflash paralysiert wird. Auf jeden Fall können wir die time-of-flight-Spektroskopie bis auf etwa 30 MeV treiben mit einem Flugweg von 400 m.

EIN TARGET FÜR HOCHSTROM-BESTRAHLUNGEN IM ZYKLOTRON

F. SCHULZ und H. BELLEMANN

Kernforschungszentrum Karlsruhe, Zyklotron-Laboratorium

A TARGET FOR HIGH-ENERGY IRRADIATIONS IN THE CYCLOTRON

ABSTRACT

A target was developed in the Karlsruhe-cyclotron which is suitable for high-energy irradiations on the inner beam. In this device the substance to be irradiated lies in the cooling water. The target can be exposed to currents of up to 100 μ A at 50 MeV (deuterons). For the production of higher neutron fluxes beryllium is used as the target material.

1. Einleitung

Um die Herstellung langlebiger Isotope am Karlsruher Zyklotron in kürzerer Zeit möglich zu machen, wurde ein sogenanntes Wassertarget für den internen Teilchenstrahl entwickelt. In dieser Targethalterung (Bild 1 und 2) liegen wasserbeständige oder wasserdicht eingepackte Targets während der Bestrahlung direkt im Kühlwasser. Man erreicht eine gute Kühlung des Targets, muß aber eine Reduzierung der maximal möglichen Teilchen-Energie in Kauf nehmen, da die Teilchen - bevor sie auf das Target treffen - zunächst ein Wolframfenster und eine dünne Kühlwasserschicht passieren müssen. Der Energieverlust beträgt bei 50 MeV Deuteronen 4 MeV, bei 100 MeV α -Teilchen 8,5 MeV.

Das Wassertarget kann Wärmebelastungen durch den Deuteronenstrahl bis zu 5 kW bei einer Leistungsdichte von 50 kW/cm^2 aufnehmen, ohne daß eine Undichtigkeit oder Beschädigung eines Teiles erfolgt.

2. Beschreibung des Targets

Das Wassertarget besteht aus zwei Hauptteilen, die aus mehreren Einzelteilen zusammengesetzt sind. Der eine Teil ist die Vakuumabschlußhaube (Teil 2,3,5 und 8 in Bild 1). Eine $50 \mu\text{m}$ dicke Wolframfolie, die U-förmig gebogen und zwischen Teil 3 und 5 eingelötet wird, dient als Eintrittsfenster für den Strahl. Die Haube wird nach mehreren Bestrahlungen so radioaktiv (1 - 2 r/h nach mehreren Tagen Abklingzeit), daß sie mit Manipulatoren gehandhabt werden muß.

Wassergekühltes Target

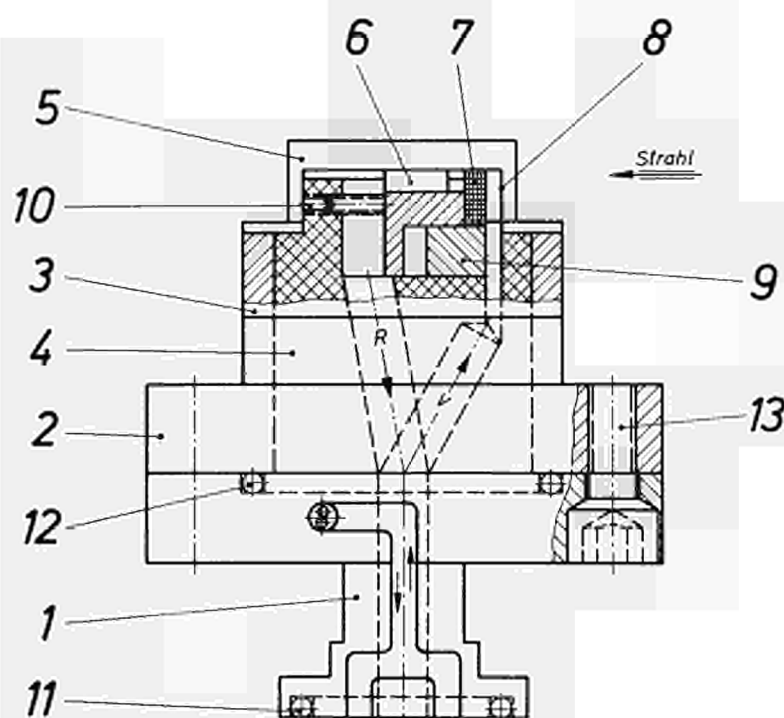


Fig. 1 Querschnitt durch das Target

1 Aluminiumfuß; 2, 3, 5 Teile der Vakuum-Abschlußhaube (Cu);
4 Kunststoffsockel; 6 Spannstück zur Befestigung des Target-
materials; 7 Targetmaterial; 8 Wolframfolie; 10 Madenschraube
zur Befestigung des Targetmaterials; 11, 12 Dichtringe;
13 Befestigungsschraube der Vakuum-Abschlußhaube.

Neutronentarget -wassergekühlt

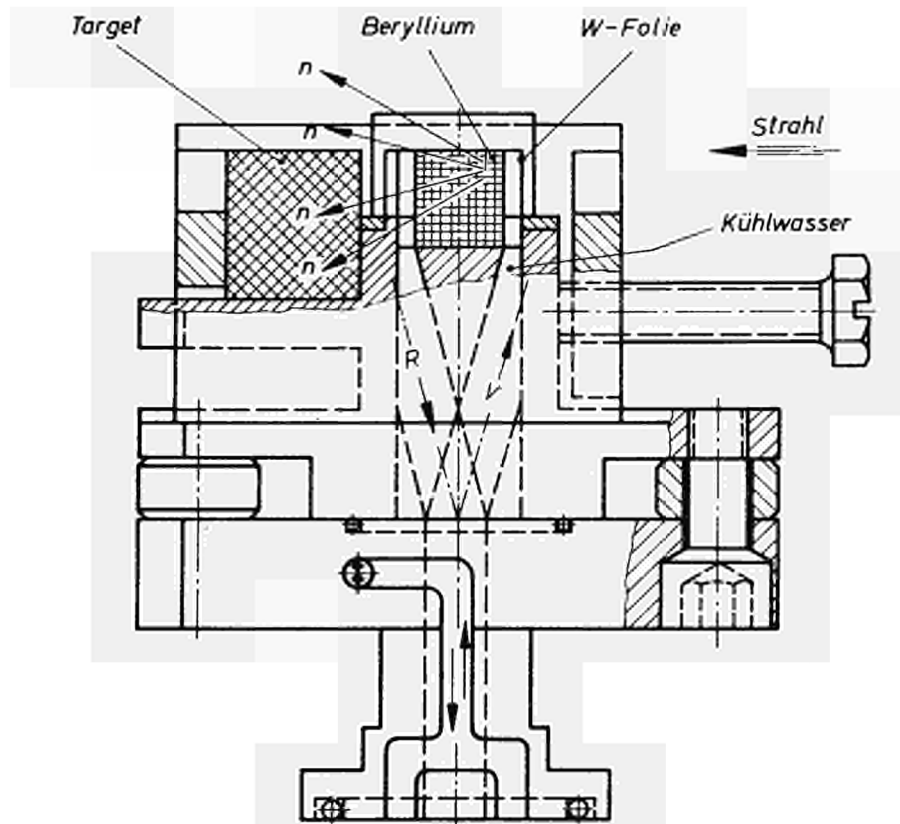


Fig. 2 Querschnitt durch die Targetanordnung zur Erzeugung von Neutronen.
Mit Target ist die Substanz bezeichnet, die mit schnellen Neutronen
aktiviert werden soll.

Der andere Hauptteil ist die eigentliche Targethalterung. Sie besteht aus einem Aluminiumfuß, der den Manipulatoren, Aufsteckschuhen und der Transportbahn angepaßt ist. Darauf ist ein Kunststoffsockel (Teil 4) aufgeklebt, in dem die Kühlbohrungen verlaufen. Das Target (Teil 7) wird mit Hilfe eines Spannstücks (Teil 6) gegen ein eingeschraubtes und verklebtes Kupferfenster gepreßt. Spannstück und Fenster sind so geformt, daß das Kühlwasser das Target umspülen muß und eine Kühlung von allen Seiten gesichert ist. Zur Entnahme des bestrahlten Targets wird mit Hilfe eines Drehmanipulators die Madenschraube (Teil 10) gelöst. Das Target fällt dann in eine bereitgestellte Schale und wird mit einem Schlitten aus der Ankunftszone ausgeschleust. Dann kann die Targethalterung ohne große Vorsichtsmaßnahmen zur evtl. erforderlichen Dekontamination oder zum Einsetzen eines neuen Targets gegeben werden. Die Targethalterung wird nicht vom Strahl getroffen und ist durch Neutronen nur schwach aktiviert. Maximal beträgt die Strahlung 50 mr/h an der Oberfläche nach einigen Tagen. Im Rücklauf des Kühlwassers befindet sich ein Thermoelement, das mit dem Kunststoffsockel vergossen ist.

3. Herstellung

Die Herstellung eines Wassertargets stellt einige Anforderungen, da das Wolframfenster (eine 50 µm dicke reine Wolframfolie) vakuumdicht zwischen Teil 3 und 5, die aus Elektrolytkupfer bestehen, eingelötet werden muß. Die Folie wird mit zwei Schrauben so eingespannt, daß Lötspalte von 0,2 mm Breite entstehen. In diese Spalte wird das Lot in Form von Blech eingeschoben, und zwar jeweils zwischen Wolfram und Kupfer. Das Vakuumlot ist eine Legierung von 72 % Ag, 20 % Cu und 8 % Ti. Seine Arbeitstemperatur von 780° liegt um 300° C unter dem Schmelzpunkt von Kupfer. Das Lot ist sehr geeignet, da es das Wolfram nur an der Lötstelle benetzt und die spätere Strahlaufftreff-Fläche vom Lot freigehalten werden kann.

Nach dem Lötprozeß im Vakuum ($< 10^{-4}$ Torr) läßt man den Teil im Vakuumofen erkalten, um eine Oxydation zu vermeiden. Nach dem Erkalten ist die Wolframfolie durch die verschiedenen Ausdehnungskoeffizienten von W und Cu leicht gewellt. Dies stört aber bei Bestrahlungen nicht. Der fertiggestellte Aufsatz wird dann mit einem dazu passend angefertigten Flansch auf Vakuumdichtigkeit geprüft. Dann lötet man diesen Aufsatz mit Weichlot (4 Teile Silber und 96 Teile Zinn) auf Teil 2. Das ergibt die fertige Targetkopfhaube.

Die Targethalterung besteht aus einem chemisch eloxierten Aluminium-Teil und einem aufgeklebten Polypropylenteil. Die Eloxalschicht schützt vor Korrosion durch vollentsalztes Wasser. Der Polypropylenteil ist strahlenschutztechnisch günstig, da er wenig aktiviert wird und durch Säuren leicht dekontaminiert werden kann. Polypropylen wurde wegen guter Strahlenbeständigkeit und seiner Dauertemperaturbeständigkeit bis zu 100°C gewählt. Es nimmt praktisch kein Wasser auf, quillt daher nicht und kann auf leichten Gleitsitz mit der Targetkopfhaube bearbeitet werden. Als Thermoelement wurde eine NiCr-Ni-Anordnung gewählt ($4,1 \text{ mV}$ bei $100^{\circ}\text{C} \pm 3^{\circ}$).

Bei Belastungsversuchen für das Strahleintrittsfenster wurden mehrere Materialien getestet: Cu, Ta, Pt, Ag, Au, Cu-Be und W. Eine $200\text{ }\mu\text{m}$ dicke Kupferfolie kann bei 50 MeV Deuteronen bis zu $40\text{ }\mu\text{A}$ belastet werden (20 kW/cm^2), eine $50\text{ }\mu\text{m}$ dicke W-Folie bis zu $100\text{ }\mu\text{A}$ (50 kW/cm^2). Die anderen Metalle waren ungeeignet. Die Kupferfolie oxydiert im Wasser auch bei geringen Wärmebelastungen. Die Folie wird infolge der auftretenden Kühlverluste zerstört, also auch bei sehr langen Bestrahlungen mit geringem Strahlstrom. Die Lebensdauer ist aber immer $1000\text{ }\mu\text{Ah}$. Die Wolframfolie oxydiert bei den angegebenen Belastungen und genügender Kühlung kaum, ist aber gegen mechanische Belastungen sehr empfindlich. Über die Lebensdauer können wir noch keine Aussagen machen, sie ist aber mindestens gleich groß. In Zukunft soll bei uns noch eine $50\text{ }\mu\text{m}$ dicke Iridiumfolie getestet werden.

4. Betriebliche Einzelheiten

Das Wassertarget wird mit Hilfe einer Kupplung während einer Bestrahlung isoliert (ca. $2 \cdot 10^8 \Omega$ gegen Masse) in der Bestrahlungskammer gehalten, so daß die Strahlstrommessung mit einem μA -Meter erfolgen kann. Die entstehenden Sekundärelektronen werden mit dem Rahmen (Teil 5) wieder eingefangen und bei der Strommessung kompensiert. Vergleichsmessungen mit Meßtargets haben ergeben, daß die Strahlstrommessung im allgemeinen bis 1 % genau erfolgen kann. Das vollentsalzte Kühlwasser wird über 2 m lange Kunststoffschläuche an das Target herangeführt. Die Leitfähigkeit des Wassers beträgt zu Beginn der Bestrahlung 1 - max. $10 \mu\text{S}/\text{cm}$. Durch Reaktionen des Targetmaterials mit dem Kühlwasser kann die Leitfähigkeit während der Bestrahlung stark zunehmen. Das Wasser wird während einer Bestrahlung stark radioaktiv und muß nach mehreren Bestrahlungen gewechselt werden. Aus diesem Grund wurden die Kühlwasserpumpe (Förderung 6 l/min bei 4 atü Staudruck) und der Rückkühler in einer wasserdichten Wanne installiert. Außerdem verlegten wir alle Ventile und Rohrverbindungsstücke in dieser Wanne, damit auftretendes Leckwasser aufgefangen wird. Nach jeder Bestrahlung wird das Kühlwasser aus dem Targetkopf in den Rückkühler ausgeblasen. Um bei einer Zerstörung der Wolframfolie zu vermeiden, daß das Zyklotron mit Wasser geflutet wird, bauten wir Magnetventile in Vor- und Rücklauf ein, die mit einem Vakuumwächter geschaltet werden (Schaltpunkt 5×10^{-3} Torr). Selbst nach einem Platzen der Folie gelangen nur wenige Tropfen Wasser in die Vakuumkammer, da der Riß sofort nach dem Druckabfall des Kühlwassers im Vakuum zueist. Das Target wird dann sofort aus der Vakuumkammer ausgeschleust, und man erhält nach einigen Minuten gutes Vakuum.

Bei einer Bestrahlung im Zyklotron im Wassertarget ist die kritische Stelle bei der entstehenden Wärme häufig nicht die dünne Vakuumabschlußfolie, sondern das eingespannte Target. Bei einer Bestrahlung von Tantal mit $50 \mu\text{A}$ Deuteronen bei 40 MeV war das Target nach 1,5 h im Strahlprofil leicht angeschmolzen,

Wasserkühlung für Beryllium-Einsatz

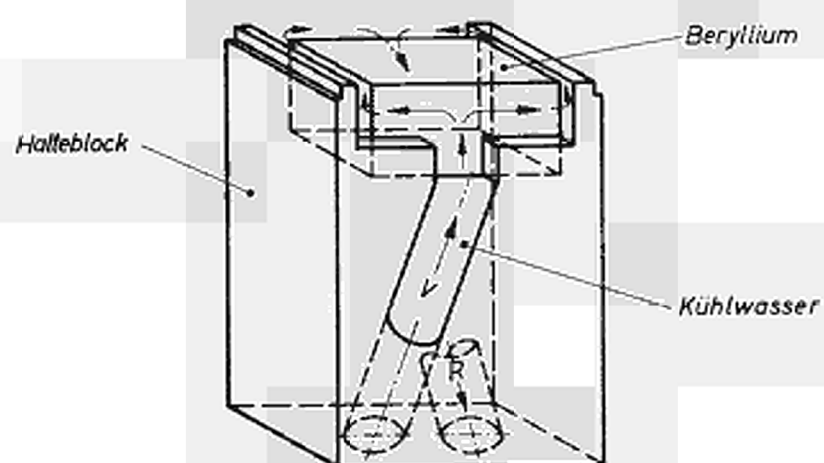


Fig. 3 Perspektivische Darstellung des Kühlwasserflusses.

obwohl das Metall unmittelbar im Wasser lag. Die Wolframfolie und alle übrigen Teile des Wassertargets waren unverändert. Solche Vorkommnisse lassen sich schlecht vermeiden, da Belastungsversuche im Wassertarget sehr zeitraubend sind. Nach jeder Probestrahlung (ca. 5 min) muß die Vakuumabschlußhaube mit einem Manipulator demontiert und das Target kontrolliert werden. Bei uns laufen Versuche, um die Belastbarkeit der Targets mittels des Thermoelements im Kühlwasserrücklauf 15 mm hinter dem Target feststellen zu können.

DISCUSSION

Mr. PAIC

By what is the 100 μ A figure for the current limited, the cyclotron operation or the target ?

Mr. SCHULZ

Es dürfte die obere Grenze sein, da die Möglichkeit der mechanischen Belastung der W-Folie voll ausgenutzt ist, wir haben ungefähr einen Staudruck von 2 atü hinter der W-Folie, das ergibt gegenüber dem Vakuum 3 kg/cm^2 Belastung. Ausserdem kann man die möglichen 100 μ A bei 50 MeV nicht ausnutzen, da die zu aktivierenden Substanzen diese Belastung nicht aushalten.

Mr. PAIC

What is the yield of neutrons ?

Mr. SCHULZ

Die möglichen Neutronenflüsse sind noch nicht gemessen, ich kann also im Augenblick nur einen Schätzwert angeben. Da man ca. 1/10 der Menge Teilchen, die man hineinschiesst, an Neutronen erhält, dürfte der Fluss ca. $10^{14} \text{ n/cm}^2/\text{sec}$ sein, direkt an der Einspannstelle für die **zu aktivierende Substanz**.

Erfahrungen in der Erzeugung hoher Flüsse an 14 MeV-Neutronen
mit Hilfe der D-T-Reaktion

G. LUTHARDT

Institut für Anorganische Chemie und Kernchemie - Universität
Mainz - Deutschland

Experience acquired in the generation of high fluxes
on 14 MeV neutrons by the D-T reaction

ABSTRACT

It is shown that in 14 MeV neutrons fluxes of $7 \cdot 10^{10}$ neutrons/cm²/sec can be created in the irradiation position using Al vapour-deposited targets containing 20 C tritium/inch² with accelerator voltages of 500-600 kV and deuteron currents of 1000-1200 μ A.

These high fluxes are measured as follows:

1. By means of the Cu (n,2n) reaction in calibrated beta and gamma counters.
Irradiation time one minute.
2. By means of the Al (n, α) Na²⁴ reaction in a 4 π - $\beta\gamma$ coincidence unit.
Irradiation time five minutes.

The half-lives of the targets used vary from 3 to 15 hours according to the target currents.

Einführung

Der Beschleuniger des Instituts für Anorganische Chemie und Kernchemie der Universität Mainz ist ein Drucktankbeschleuniger vom Cockcroft-Walton-Typ, gebaut von der Fa. Philips, Eindhoven, und inzwischen fast 10 Jahre alt. Er ist ausgelegt für eine maximale Spannung von 2 MV, mit der aber bisher nicht gearbeitet wurde. Beschleunigt werden Deuterium-Ionen, die in einer Hochfrequenz-Ionenquelle mit induktiver Kopplung erzeugt werden. Die früher sehr viel verwendeten Beryllium-Targets wurden auf Kupferscheiben aufgelötet und diese mit Wasser direkt gekühlt. Die Tritium-Targets werden in eine Halterung aus Aluminium eingesetzt, die Ränder mit Araldit, einem Epoxidharz, gekittet und ebenfalls direkt mit Wasser gekühlt. Der Abstand Brennfleck - Bestrahlungsprobe wird auf diese Weise sehr klein gehalten und beträgt etwa 5 mm.

Was ist unter hohen Flüssen zu verstehen?

Was verstehen wir nun unter hohen Flüssen an 14 MeV-Neutronen? Das soll in einer kurzen Schilderung der Entwicklung bis zum gegenwärtigen Stand aufgezeigt werden:

Anfangs wurden Targets von etwa 1 cm \varnothing und einem Tritium-Gehalt von 1 Ci verwendet. Von diesen Targets liegen keine exakten Messungen weder über die Flüsse noch über den Leistungsabfall vor. Ungefähre Schätzungen geben einen Fluß von etwa $10^9 \text{ n/cm}^2 \text{ sec}$ und eine Halbwertszeit des Targets von 1 - 1,5 Std.

Diese Targets wurden mit 250 kV-Beschleunigungsspannung und 100 μ A Targetstrom gefahren.

Mit 4 Ci-Targets wurden dann schon Flüsse von $7-8 \times 10^9 \text{ n/cm}^2 \text{ sec}$ bei 300 kV Spannung und 200 - 250 μ A Strom erreicht. Die Halbwertszeit beträgt bei diesen Targets etwa 8 - 10 Std. und sie stellten einen beträchtlichen Fortschritt dar.

Targets mit einem Tritium-Gehalt von 20 Ci/inch^2 , die der größeren Schichtdicke wegen mit 350 kV Spannung und einem Strom von 200 - 250 μ A gefahren wurden, ergeben bei diesem Strom einen anfänglichen Fluß von $7 \times 10^9 \text{ n/cm}^2 \text{ sec}$ und eine Halbwertszeit von etwa 17 Std., die sich dann nach längerer Benutzung noch erhöht. Wie lange die Halbwertszeit bei Flüssen um $1 - 2 \times 10^9 \text{ n/cm}^2 \text{ sec}$ wird, wurde nicht gemessen, da Flüsse in diesem Bereich nicht mehr gefragt sind. Um anstehende Probleme zu lösen, wurde versucht, die Flüsse höher und höher zu treiben. Dies wird durch eine Erhöhung des Targetstromes erreicht. Gleichzeitig wurden die inzwischen kommerziell erhältlichen Al-bedampften Targets eingesetzt, die bei höherer Spannung gefahren und so thermisch höher belastet werden. Für ein 5 Ci-Target wurde eine Halbwertszeit von 7 - 8 Std. bei 400 μ A Strom gemessen. Die anfänglichen Flüsse lagen bei $1,5 \times 10^{10} \text{ n/cm}^2 \text{ sec}$.

In der Folgezeit wurden ausnahmslos Al-bedampfte Targets mit $20 \text{ Ci-Tritium/inch}^2$ verwendet. Die erreichten Flüsse stiegen auf etwa $3 \times 10^{10} \text{ n/cm}^2 \text{ sec}$ für 350 kV-Beschleunigungs-

spannung und 400 μA Strom. Die Neutronenausbeute fällt ab mit etwa 9 h Halbwertszeit. Doch von nun an wird es schwierig, Targethalbwegszeiten anzugeben, weil für die meist kurzen Bestrahlungszeiten von einigen Sekunden bis zu mehreren Minuten höhere Ströme verwendet wurden. Inzwischen werden Ströme von 1000 - 1.200 μA erhalten. Dazwischen werden dann die Targets für Bestrahlungen im Stundenbereich mit geringeren Strömen gefahren. Es ergeben sich unter diesen sehr gemischten Bedingungen Targethalbwegszeiten im Bereich von 8-10 Std. Ein Target wurde konstant mit Strömen von 900 - 1000 μA gefahren, die Halbwertszeit beträgt hierbei etwa 4,5 Std.

Doch der Wunsch nach höheren Flüssen ist bei den meisten Benutzern noch immer vorhanden. Im Augenblick werden in unserem Hause mit neuen 20 Ci-Tritium-Targets Flüsse von maximal $7 \times 10^{10} \text{ n/cm}^2 \text{ sec}$, gemessen über die $\text{Cu}(n,2n)$ -Reaktion, erreicht. Der Strom liegt bei 1.200 μA , die Spannung, wie jetzt bei allen Al-bedampften Targets, zwischen 500 und 600 kV je nach Target. Die Spannung wird so festgelegt, daß bei einem niedrigen Targetstrom mit einem fest installierten Neutronenzähler über dessen Zählrate die Beschleunigungsspannung ermittelt wird, die die höchste Neutronenausbeute ergibt. Mit dieser Spannung wird dann das jeweilige Target in der Folgezeit gefahren. Die Halbwertszeit der Targets beträgt trotz der recht hohen thermischen Belastung von etwa 600 - 700 Watt etwa 3 Std. Die Benutzer hoffen, daß in der Zukunft noch weitere Verbesserungen möglich sein werden.

Die Meßmethoden

Die Neutronenflüsse werden mithilfe der $\text{Cu}(n,2n)$ -Reaktion bestimmt. Verwendet werden Kupfer-Folien von etwa 7,5 mg Gewicht, die in einem geeichten Methan-Durchflußproportionalzähler gemessen werden. Durch die Erzeugung höherer Flüsse wurde das Auflösungsvermögen des Zählrohres bei weitem überschritten. Daher werden jetzt Folien von etwa 2 mg Gewicht eingesetzt. Die erhaltenen Anfangsaktivitäten für eine Minute Bestrahlungsdauer liegen bei 650.000 Imp/min, der entsprechende Fluß beträgt $6 - 7 \times 10^{10} \text{ n/cm}^2 \text{ sec}$. Doch dieser Fluß herrscht natürlich nur an der Stelle des Targets, die unmittelbar vom Deuteronenstrahl getroffen wird. Dieser Brennfleck hat eine Größe von etwa 1 cm^2 und kann leider nur durch kleine Proben ausgenutzt werden.

Um einen direkten Vergleich mit anderen Laboratorien zu haben, wollten auch wir in Mainz uns der Texas-Convention anschließen, wie sie 1965 auf der Konferenz "Modern Trends in Activation Analysis" in College Station vorgeschlagen wurde. In einem Anhang zu dieser Convention schlägt Heath vor, genormte Kupfer-Scheibchen von 0,25 mm Dicke und 1 cm bzw. 2,5 cm \varnothing zu benutzen und die Cu 62-Aktivität mit einem 3 x 3 inch NaJ-Kristall unter Standardbedingungen zu messen (1). Doch selbst mit der kleinen Cu-Scheibe von 1 cm \varnothing , 0,25 mm Dicke, und einer Minute Bestrahlungsdauer ist die erhaltene Aktivität viel zu hoch. Noch 15 Minuten nach Bestrahlungsende ist die Totzeit des benutzten Spektrometers größer als 70 %!

Daher wurden auch hier wieder die Kupfer-Folien von 7,5 mg Gewicht eingesetzt. Die erhaltenen Daten sind nach einer Eichkurve von Mundschenk, der einen 3 x 3-Zoll-Kristall mit Hilfe von Nukliden, die durch 4π - β - γ Koinzidenzmessungen absolut gezählt wurden, geeicht hat, auf Absolutaktivität umgerechnet (2). Die Ergebnisse sind in sehr guter Übereinstimmung mit den nach Heath, IDO-Report 16880 (1964) korrigierten Daten (3). Ein Unsicherheitsfaktor in diesen Berechnungen war die nicht exakt bekannte Dicke der Kristallumhüllung, die mit 0,7 cm angenommen wurde, um dann aus den Heath'schen Kurven extrapolieren zu können.

Die durch die Gammamessungen erhaltenen Flußwerte stimmen sehr gut mit den durch die Betamessungen erzielten überein. Doch auch diese gelten nur für den Brennfleck des Targets. Viele unserer Benutzer haben beträchtlich größere Proben mit einer Fläche von etwa 5 cm² entsprechend der Bohrung unserer Targethalterung, in der Mengen von etwa 5 - 6 g Uranylнитrat bestrahlt werden können. Diese Benutzer interessiert der Fluß, den die gesamte Probe erhalten hat, also ein über die Fläche von etwa 5 cm² gemittelter Fluß. Um diesen bestimmen zu können, wurde die Reaktion $\text{Al}(n, \alpha) \text{Na } 24$ ausgenutzt. Die eingesetzten Al-Folien haben ein Gewicht von etwa 5 mg/cm² bei einer Fläche von 5 cm². Die induzierte Na 24-Aktivität wird mithilfe der 4π - β - γ -Koinzidenz-Methode absolut gezählt, die, wie Menke und Fahland in unserem Institut gezeigt haben, auch für dicke Proben anwendbar ist (4). Angenommen wurde ein

Wirkungsquerschnitt von 115 mbarn für diese Reaktion. Jetzt errechnen sich Flüsse pro cm^2 , die etwa um den Faktor 2,3 kleiner sind als die mithilfe der $\text{Cu}(n,2n)$ -Reaktion erhaltenen, aber für die gesamte Probe gelten. Der höchste über diese Reaktion gemessene Fluß war $4,1 \times 10^{10} \text{ n/cm}^2 \text{ sec}$.

Auch für diese Werte liegt eine Bestätigung aufgrund einer anderen Reaktion vor. Denig und Trautmann (5) aus unserem Institut bestimmten den Wirkungsquerschnitt der Reaktion $^{238}\text{U}(n,p)^{238}\text{Pa}$ über die Aktivität des Spaltproduktes $\text{Ba } 140$, das ja gleichzeitig in der Probe entsteht. Die Bestrahlungsprobe hatte die Größe von etwa 5 cm^2 und bestand aus 1 g Uran als Uranylнитрат.

Alle diese Ergebnisse wurden mit der Zählrate des mitlaufenden, in etwa 6 m Entfernung vom Target festinstallierten Neutronenzählers mit einem BF_3 -Zählrohr verglichen. Die Ergebnisse zeigen die Proportionalität zwischen gemessenem Neutronenfluß und Zählrate. So kann in Zukunft der Neutronenfluß, vor allem dessen zeitliche Veränderung bei längeren Bestrahlungen, über die Zählrate des Neutronenzählers direkt abgelesen werden. Nur gelegentlich werden direkte Messungen nötig sein, um die Funktion des Zählrohres zu überprüfen. Das kann auch über die Messung einer Ra-Be-Neutronen-Quelle als Neutronenstandard erfolgen, die in einer definierten Position zum Zählrohr eingebracht werden kann.

Zum Abschluß möchte ich noch auf einige neuere Ergebnisse aus unserem Institut eingehen, die erst durch solche hohen Flüsse
Neutronen
an 14 MeV-erzielt werden konnten.

Die bereits erwähnte Reaktion $U\ 238\ (n,p)\ Pa\ 238$, Halbwertszeit 2,3 min, hat einen Wirkungsquerschnitt von etwa 1 mbarn, aber die selektive Abtrennung des Protaktiniums von 1 g Uranyl-salzen und den gleichzeitig entstandenen Spaltprodukten hat eine chemische Ausbeute von nur 10 - 15 %. Daher mußten mehrere Versuche addiert werden, um Daten über das Zerfalls-Schema dieses neuen Nuklides mit der notwendigen Statistik erhalten zu können.

Ein weiteres Beispiel liefert die Untersuchung der Reaktion

$Dy\ 164\ (n,p)\ Tb\ 164$, mit dem Zweck, das komplexe Zerfalls-Schema dieses deformierten Kernes aufzuklären. Der Wirkungsquerschnitt beträgt etwa 2 - 3 mbarn, die Halbwertszeit etwa 3 min. Die Aufnahme des Gamma-Spektrums mit einem Ge-Li-Detektor, vor allem Aufnahmen von Gamma-Gamma-Koinzidenz-Spektren machten trotz des hohen Flusses Aufsummierungen von 20 - 25 Einzelversuchen nötig (6).

Es soll noch erwähnt werden, daß im Jahre 1965 insgesamt 1.131 Versuche mit einer Gesamtbestrahlungsdauer von 392 Std. mit Tritiumtargets durchgeführt wurden. 1966 waren es bereits 1.379 Versuche und einer Gesamtdauer von 306 Std.

Doch das Problem der Erzeugung hoher Flüsse an 14 MeV-Neutronen muß auch unter finanziellem Gesichtspunkt betrachtet werden.

Die Frage: ist es sinnvoll ein Target mit einem Wert von etwa

900,-- DM in wenigen Stunden auszubrennen, muß aber unbedingt bejaht werden. Denn die Alternative lautet in diesen Fällen, entweder noch mehr Einzelversuche aufzusummieren und wertvolle Meßgeräte, wie z. B. ein 4096-Kanal-Spektrometer, längere Zeit zu blockieren oder noch größere Mengen an angereicherten stabilen Isotopen zu kaufen, die im speziellen Falle zwischen 0,2 - 1,5 Dollar pro mg kosten.

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DISCUSSION

Mr. LAVERLOCHERE

Quelles conditions expérimentales avez vous utilisées pour les mesures selon la Convention Texas ? A quelle distance de la cible avez-vous placé le disque de cuivre ? La Convention Texas n'impose pas de distance, afin que chaque expérimentateur puisse l'adapter à la puissance de la source de neutrons. Je suis donc surpris des difficultés que vous semblez avoir eues vis à vis de la trop forte activité de ces disques de cuivre?

Mr. LUTHARDT

Die Kupferscheibe, die den Bedingungen der " Texas Convention " entspricht, wurde in der Bohrung der Targethalterung bestrahlt. Der Abstand zum Brennfleck beträgt etwa 5 mm. Dabei entsteht dann eine zu hohe Cu-Aktivität.

Mr. CUYPERS

Si l'activité du cuivre est trop élevée, pourquoi ne peut-on laisser décroître partiellement l'activité de celle-ci ?

Mr. LUTHARDT

Uns interessiert der Fluss in der Position der Probe. Sicherlich kann man die Kupferscheibe zur Flussbestimmung in grösserer Entfernung vom Target bestrahlen. Doch es erscheint mir einfacher, eine kleinere Cu-Menge zu benutzen, als hinterher auf den Fluss am Ort der Bestrahlungsprobe umzurechnen.

Mr. SMITH

At A.W.C.E. the aluminium (n, α) Na²⁴ reaction is used as a neutron flux monitor by users who place samples close to the neutron source. Usually a "sandwich" is made of aluminium foil / sample / aluminium foil. Although I am in a position to measure the source strength accurately but not the flux through a users sample.

Mr. RICCI

The 14 MeV neutron spectrum depends on the geometrical construction of the target; it is this very spectrum which determines the activation of samples and, particularly, the Cu monitor. Thus, I think the Texas Convention for 14-MeV neutron-flux measurement should be revised, because comparison of fluxes at two different facilities (different constructing materials and shapes) could give unrealistic results.

Mr. LAVERLOCHERE

Le cuivre a été choisi pour la Convention Texas à cause du seuil assez élevé de la réaction utilisée. Le but de cette convention est en effet de permettre des comparaisons pour des neutrons dans la zone des 14 MeV (réaction D,T), et seulement dans ce cas.

PERFORMANCE OF TARGETS IN SEALED-OFF NEUTRON TUBES

M. Hillier, P.D. Lomer, D.S. Stark, and J.D.L.H. Wood

Services Electronics Research Laboratory
Baldock, Hertfordshire, England

ABSTRACT

Intense and long-lived sources of 14 MeV neutrons may best be achieved with sealed-off generator tubes. The evolution of targets for such tubes is discussed. Lives in excess of 100 hours have already been achieved at levels of 10^{10} and 10^{11} neutrons/second. At 10^{12} neutrons/second, lives of several hundred hours are foreseen.

1. Introduction

Intense sources of 14 MeV neutrons have important applications in the fields of reactor instrumentation, activation analysis and radiotherapy. Many pumped accelerators have been constructed using the D-T reaction with yields varying from 10^9 up to 2×10^{13} neutrons/second.⁽¹⁾ However, as far as a routine user is concerned output alone is not enough; long life is of paramount importance.

The principle causes of target deterioration with time are

1. Inadequate cooling
2. Poor vacuum
3. Dilution of target tritium by deuterium from the ion beam
4. Sputtering of target material.

The use of a D-T mixture in a well-processed sealed-off tube eliminates the dilution problem and greatly improves the vacuum conditions. There remain the questions of cooling and sputtering. These factors will be discussed in detail in relation to a source designed for a yield of 10^{12} neutrons/second from a 6 cm^2 target with a life of several hundred hours.

The required deuteron currents may be obtained from Fig. 1 as 7 mA at 150 kV or 4 mA at 300 kV. In practice efficiency is reduced by a factor of up to 2 by losses due to presence of molecular ions, hydrogen impurities and surface contamination of the target, and a more realistic figure is 10 mA at 300 kV; a power of 3 kW.

2. Target Cooling

The dissociation pressure temperature curves for titanium and erbium are given in Fig. 2. The rare earth targets are much more thermally stable than titanium and if we assume a dissociation pressure of 10^{-3} torr as being an upper limit in the target region of the accelerator, this corresponds to limiting temperatures of 230°C for titanium and 600°C for erbium. The effect of this has been observed by Dubus⁽²⁾ in experiments comparing half lives of Er and Ti at high intensities.

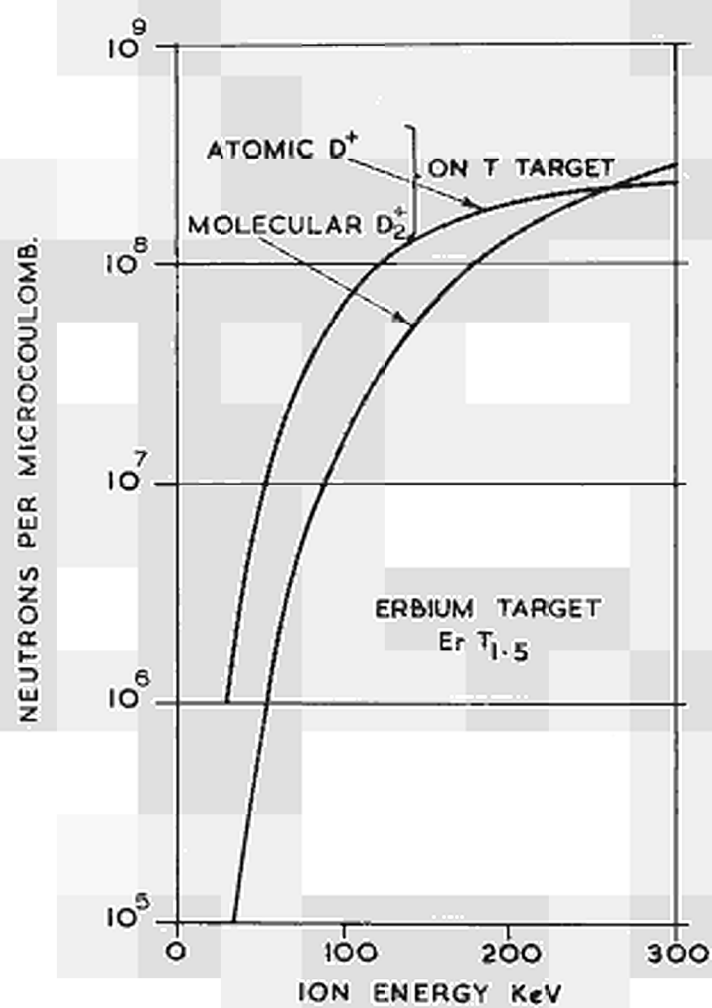


FIG I. YIELD FROM D-T REACTION.

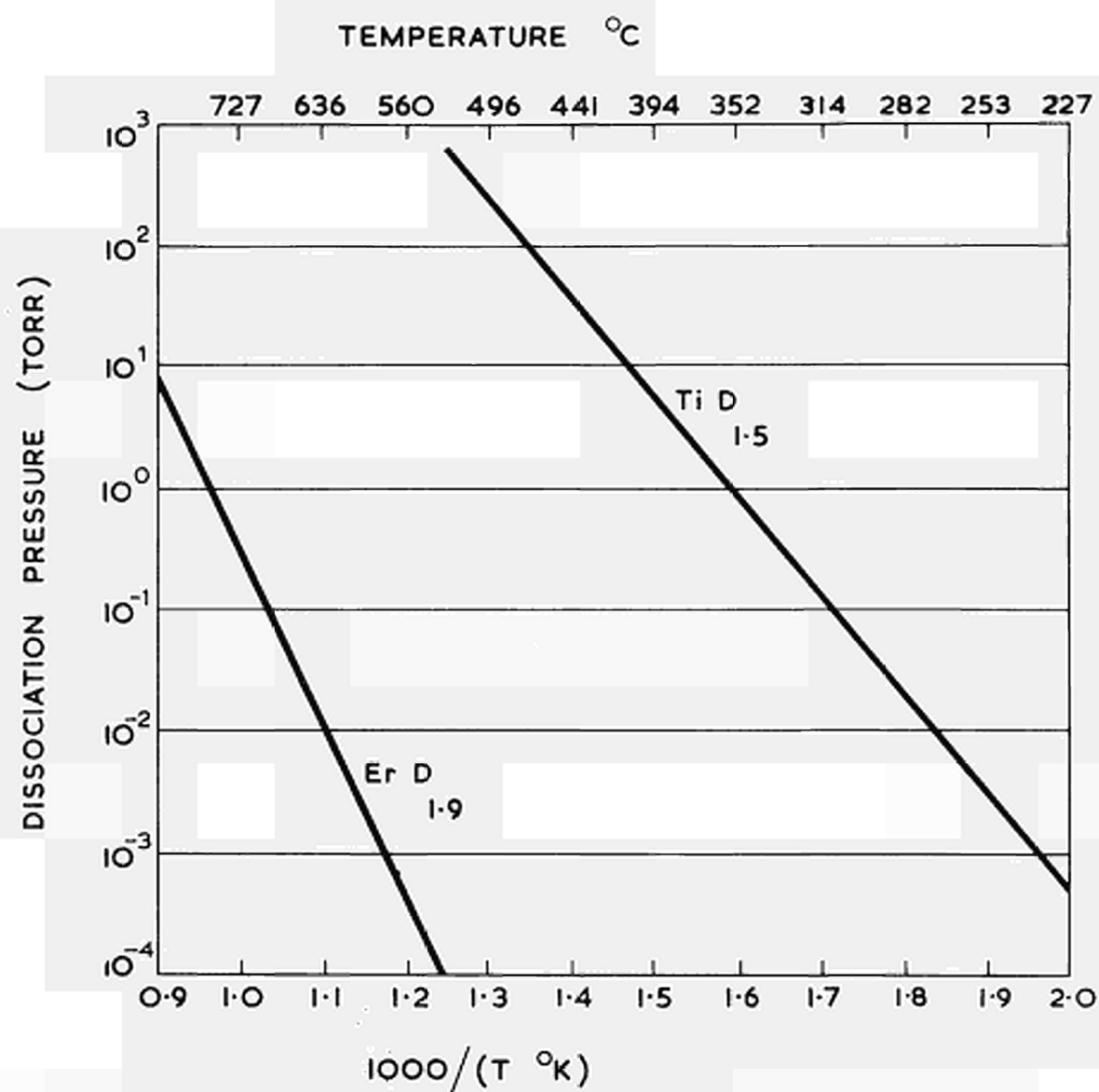


FIG 2. DISSOCIATION PRESSURE AS A FUNCTION OF TEMPERATURE FOR ERBIUM DEUTERIDE AND TITANIUM DEUTERIDE

If we consider target cooling, the temperature at the surface of the target is determined by the incident power density, the thickness of film and substrate and the velocity of cooling liquid, in this case, water. The minimum water velocity as a function of power density for a standard titanium target is given in Fig. 3, assuming that the surface temperature of the target must not exceed 200°C corresponding to 10^{-3} torr dissociation pressure. The surface temperature is given as a function of cooling velocity in Fig. 4 for a neutron output of 10^{12} neutrons/second i.e. 3 kW over a target area of 6 cm². This shows that with titanium the velocity must exceed 8 ft/sec whereas with erbium much lower velocities are acceptable. In order to keep the back surface of the target below the boiling point of water, pressurization is needed to the extent shown on the graph. Provided these cooling rates are exceeded loss of output due to thermal instability of the targets should not be a problem.

3. Vacuum Conditions

In the sealed tube, vacuum conditions are optimum and the build-up of surface films is no longer the serious problem faced by users of continuously pumped accelerators.

4. Dilution

Target dilution effects have been measured by many workers. Some typical results are shown in Table I. Most of these were reported at the 1965 meeting on Neutron Targets at Grenoble.

TABLE I

SOURCE	CONDITIONS	HALF-LIFE
Dubus ² (Grenoble)	400 kV 0.5 mA	0.8 - 1.2 mA.hr/cm ²
Morgan ³ (Texas Nuclear)	1/4 in beam spot 150 kV 300 μ A 3-5 Ci/in ²	0.5 mA.hr/cm ²
Broerse ⁴ (Rijswijk)	400 kV 150 μ A 2 cm ² 1.5 Ci/in ²	1.28 mA.hr/cm ²
Coon ⁵ (Los Alamos)	250 kV	0.6 mA.hr/cm ² (Proportional to beam energy)
Perkin ⁶ (AWRE)	300 kV 10 mA 10^{12} n/sec 650 cm ² (2000 Ci) rotating 5cm beam diameter	3 mA.hr/cm ² (static) 3 mA.hr/cm ² (rotating)

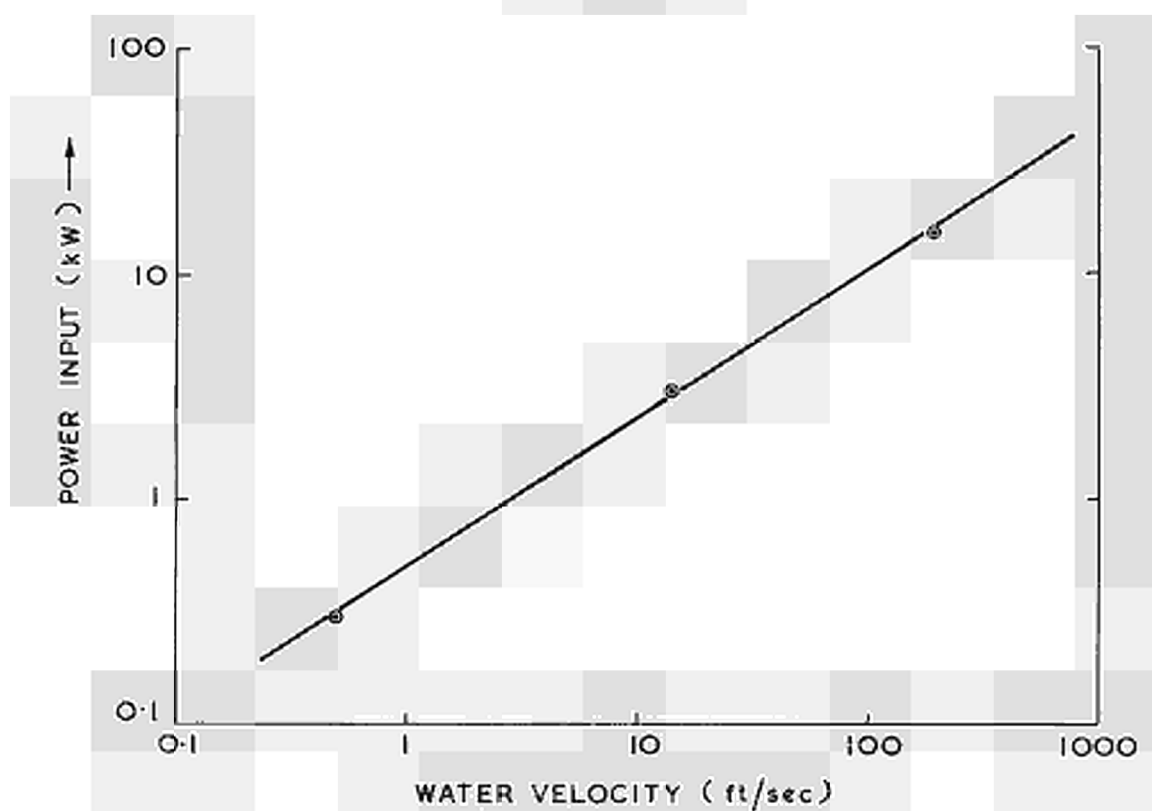


FIG 3. POWER INPUT versus COOLING WATER VELOCITY
REQUIRED FOR 200°C AT TARGET SURFACE.
(10μ Er or Ti target on 1"x1"x0.020" Mo substrate)

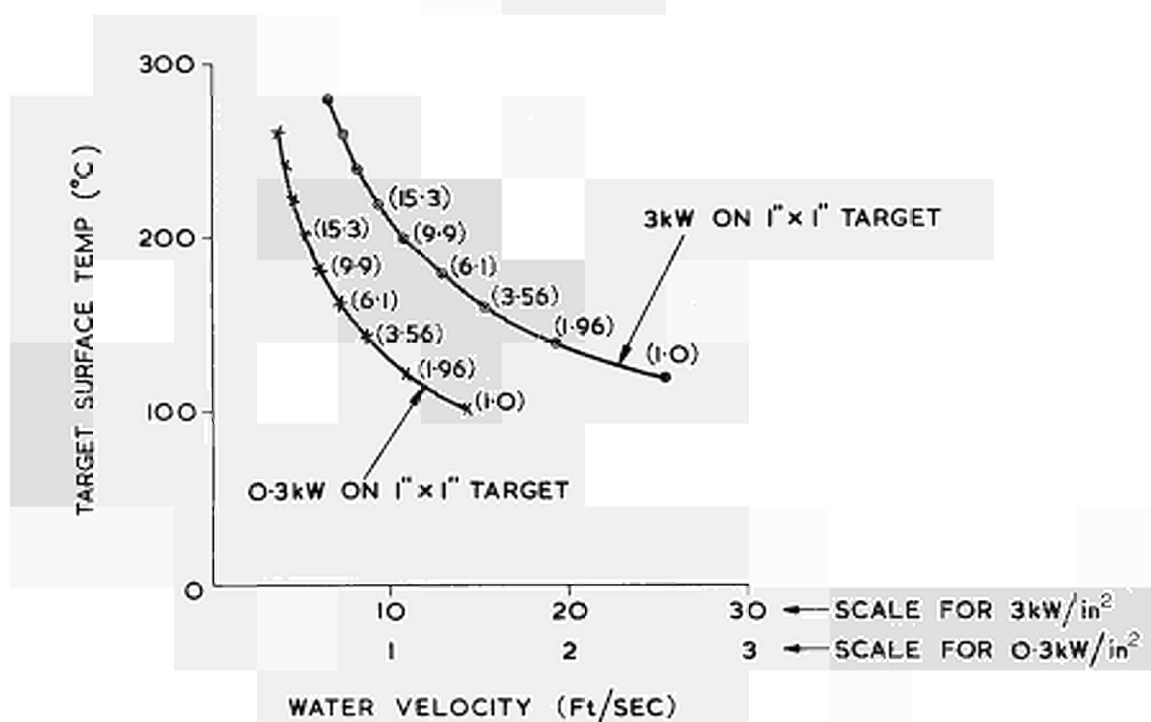


FIG 4. TARGET SURFACE TEMPERATURE VERSUS COOLING WATER VELOCITY

(figures in brackets show pressurisation of water (atmospheres)
to maintain non-boiling cooling conditions)

(10 μ Erbium or Titanium target on a 0.020" thick molybdenum
substrate. Cooling water at 20°C)

<u>SOURCE</u>	<u>CONDITIONS</u>	<u>HALF-LIFE</u>
Hollister ⁷ (Kaman)	with clean system	1 mA.hr/cm ²
Booth ⁹ (Livermore)	300 - 500 KeV 200 cm ²	2 mA.hr/cm ² (rotating)

Taking a figure of 1.0 mA.hr/cm² for half-life which fits well with a simple model of deuterons replacing tritium in the target, a neutron output of 10¹² neutrons/second requiring 10 mA at 300 kV will have the half-life due to dilution of 36 minutes - assuming a 6 cm² target. Alternatively with a rotating target of the design described by Cossuta with an area of 121 cm² a half-life of 12 hours is predicted.

5. Sputtering

The comparison of the life of a target due to dilution with that due to sputtering is shown in Fig. 5. Here we have taken the dilution life as the time taken for the number of incident deuterons to equal the number of tritium atoms in the target for a current giving 10¹² neutrons/second initially. The sputtering limit is based on data we have published for erbium which was presented at the 1965 Grenoble meeting⁽⁸⁾. A ratio of 150 is obtained at all energies and this shows that sputtering is quite insignificant in relation to dilution in considering accelerators using deuteron beams on titanium targets.

6. Use of D-T Mixtures

The use of D-T mixtures in both ion source and target eliminates the dilution problem so that we can extend target lives to those limited by sputtering. Fig. 6 shows the energy dependence of neutron output for D-T mixtures with three cases of pure atomic, pure molecular and 50% atomic beams. At higher energies, there is little advantage in having a high percentage of atomic ions. The efficiency is shown as a function of ion energy in Fig. 7 which shows that whereas with D on T the maximum efficiency is achieved at 150 keV, with D-T mixtures the efficiency reaches a maximum in the 200 - 300 keV range for beams predominant in atomic ions.

The sealed-off neutron tubes which we have developed are basically to the design of Fig. 8. The tube consists of an r.f. ion source from which a mixed beam of deuterons and tritons is accelerated on to a mixed gas target. Tubes were rigorously cleaned and processed to avoid subsequent contamination of the D-T mixture which was supplied from a heated titanium replenisher. Thus the ion beam is believed to be of high

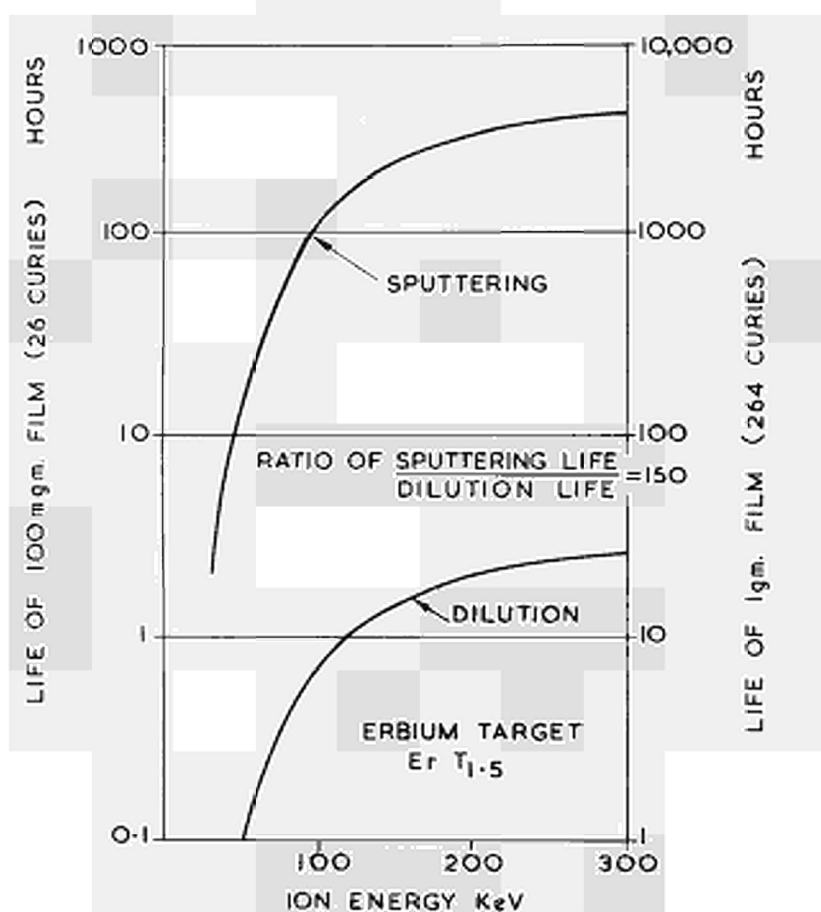


FIG 5. LIFE OF TRITIUM TARGET UNDER DEUTERON BOMBARDMENT.
(NEUTRON OUTPUT 10^{12} /SEC)

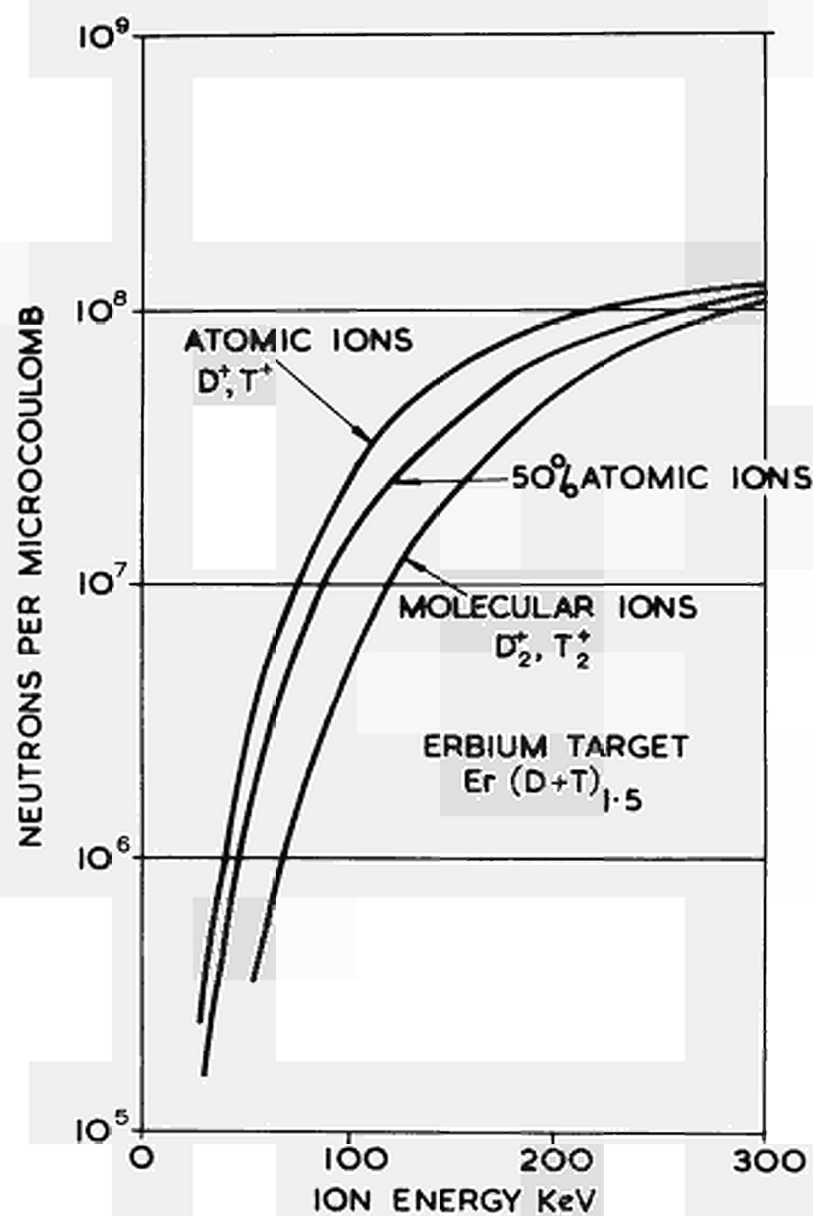


FIG 6. MIXED BEAM YIELD FROM MIXED TARGET

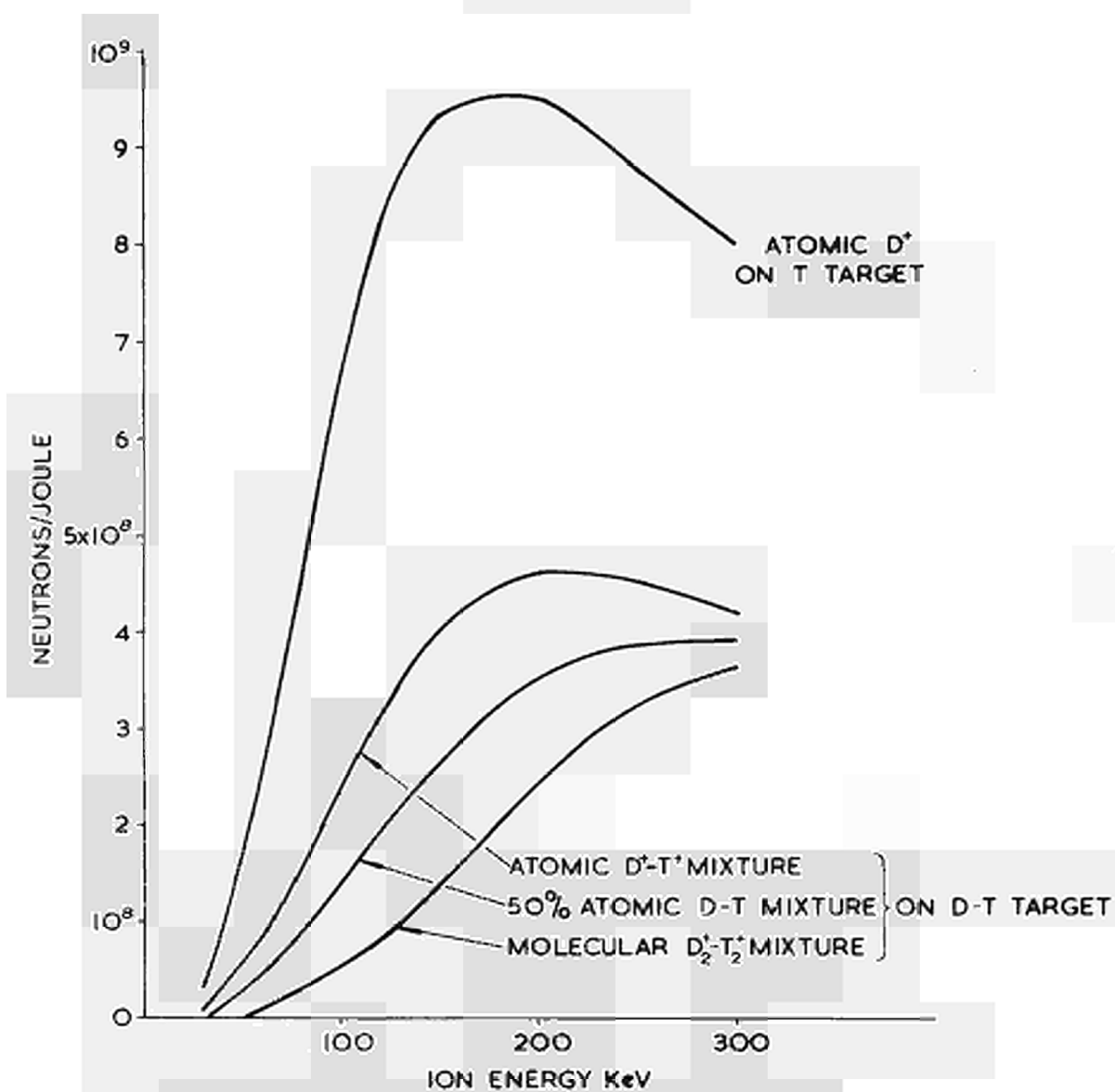


FIG 7. EFFICIENCY OF D/T REACTION

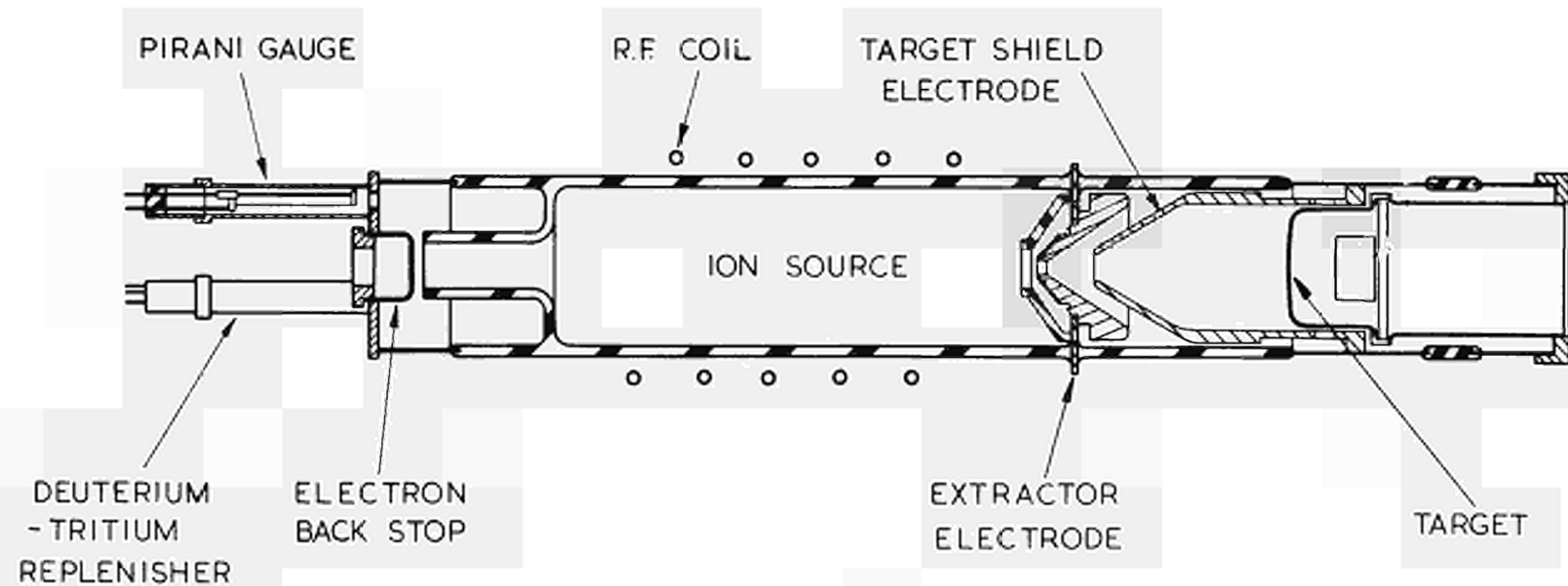


FIGURE 8. L-TUBE 10^{10} NEUTRONS/SEC.

purity with a high percentage of atomic ions. Most of the results to be presented have been obtained from this tube operating at a current density of 0.7 mA/cm^2 and an accelerating voltage of 110 kV corresponding to an output of 10^{10} neutrons/second. Some results have also been obtained at current densities of 2.5 mA/cm^2 at 120 kV with a tube producing 10^{11} neutrons/second. This tube is very similar in size and in principle to that shown in Fig. 8. The major difference lies in the addition of a beam extraction electrode necessary to achieve the ion currents of 12 mA.

Fig. 9 gives some typical results of neutron output measurements during continuous operation of such sources, using erbium targets of various thicknesses, also in one case for a scandium target. The thin targets both of erbium, 2 mg/cm^2 and scandium, $.5 \text{ mg/cm}^2$ were used in order to measure sputtering rates. On each of these curves we see an initial level period followed by a slow decline in output to another level condition. The initial flat portion corresponds to the yield for the intact target. The slope represents the gradual radial erosion of the target film by sputtering due to the non-uniform ion beam. The final flat portion is thought to correspond to the self-loaded target formed in the molybdenum substrate. Fig. 10 shows the appearance of the targets after operation. In Fig. 10 c) following 200 hours operation at 10 mA at 120 kV, the large central sputtered region can clearly be seen. The diameter of the beam is about 2.8 cm. Fig. 11 shows autoradiographs of these targets showing the tritium distribution. Fig. 12 is an autoradiograph of the back face of two targets and represents the distribution of induced activity in the molybdenum. The variation in molybdenum activity across the target substrate is directly related to the variation in neutron flux across the target. This distribution will depend on two factors; ion density variation in the beam and the extent to which the target material has been sputtered away. Such a radiograph taken early in the target's history when the film is intact will represent the ion beam distribution alone, and subsequently as the film is sputtered one gets the compound of these two effects. From a series of such radiographs taken during the life of a target it was found possible to derive a value for the sputtering coefficient as previously reported at the 1965 Grenoble Conference.⁽⁸⁾ The sputtering coefficient determined was .007 atoms per ion for 50/50 mixture deuterium and tritium, mostly

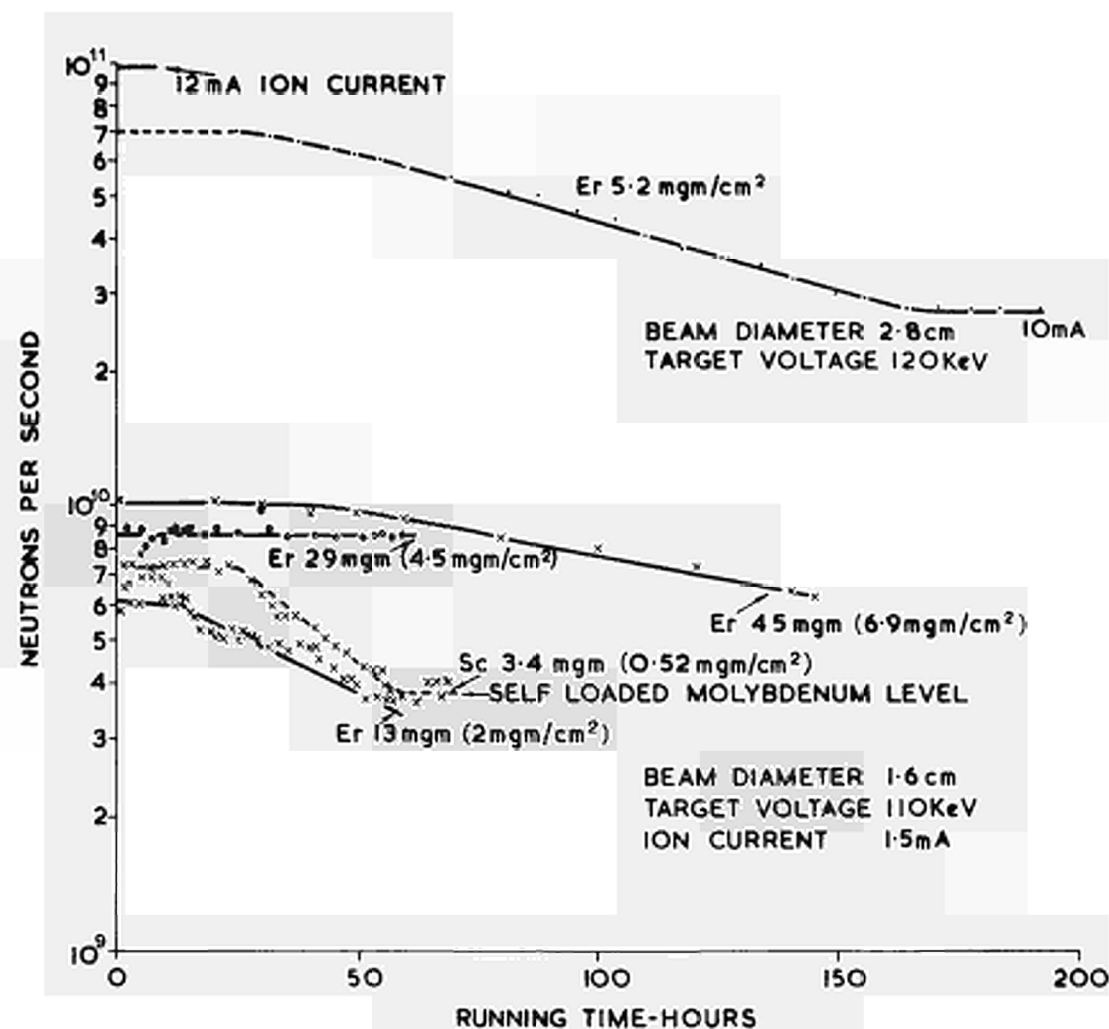


FIGURE 9. TARGET LIFE

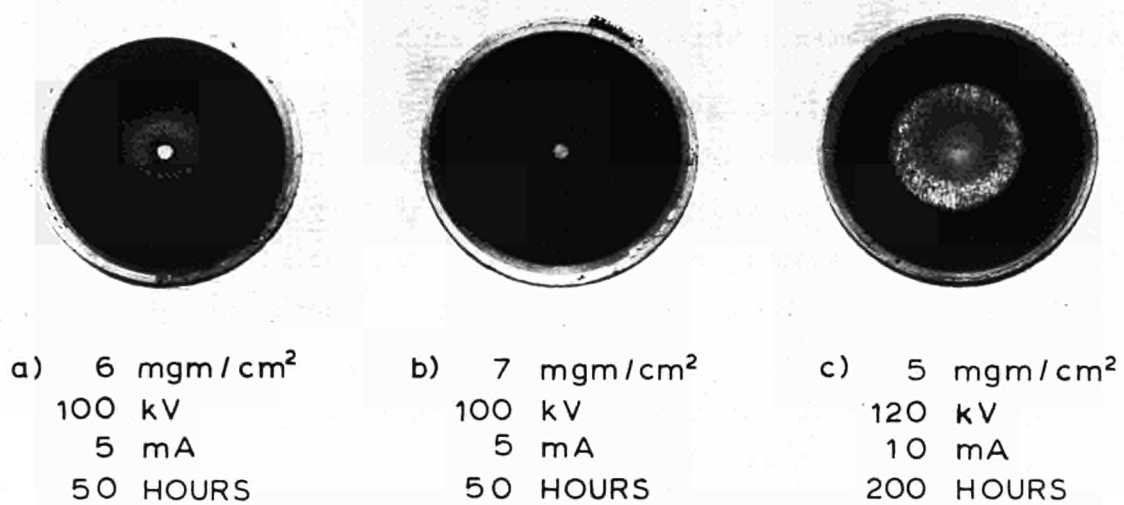


FIGURE 10. Appearance of Erbium Targets

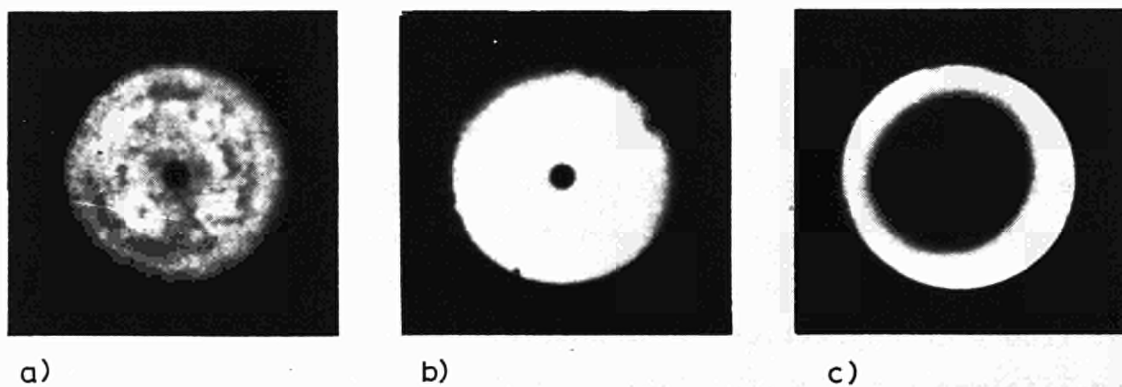


FIGURE 11. Tritium Autoradiographs
(of targets in FIG. 10)

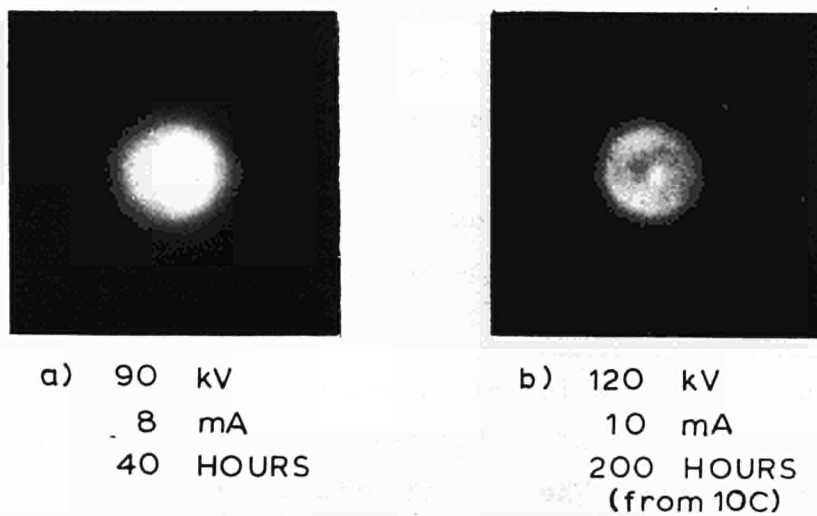


FIGURE 12. Autoradiographs of activity
in Molybdenum substrate.

atomic, impinging on an erbium target with an energy of 110 kV. A similar value was also obtained with the scandium target. The decline in output as seen on the high intensity curve of Fig. 9 is quite compatible with the measurements which were made at the low levels.

The interesting possibility that the level portions of the ends of these curves correspond to drive-in targets has been investigated using tubes containing uncoated molybdenum targets. The results are shown in Fig. 13. The neutron output increases as the surface of the molybdenum is loaded reaching a value of 2.8×10^9 neutrons/second after an hour, corresponding to the results at the end of the experiment with scandium. Similar experiments have now been started with molybdenum targets at the high current level of 10 mA. Up to 5 mA the output was found to increase linearly with current but beyond this there was a rapid fall-off presumably due to overheating of the molybdenum and consequent release of the gas. It is now proposed to investigate the effects of cooling parameters on self-load targets of this sort. The advantages to be obtained from using self-load targets are tremendous in that sputtering of an evaporated film is of course no longer a problem and extremely long lives may be obtained, which are probably only terminated by other tube parameters. Unfortunately the neutron outputs determined so far are only between one third and a half of that from a loaded erbium film. However in some cases the advantages of long-life may outweigh the lack of yield.

Our future experiments on the tube with a 12 mA beam will include a study of much thicker evaporated films at least five times thicker than those in present use, with the aim of maintaining the output constant for a period of at least 100 hours. Such targets have been evaporated and loaded successfully. Yield measurements on an accelerator have shown normal performance but a life-test in a tube has yet to be carried out. As for drive-in targets we shall continue to investigate the possibilities here with variations in cooling rate and possible other target materials.

7. Design for 10^{12} neutrons/second

The possible application of neutrons to radiotherapy has created interest in the development of 14 MeV neutron sources with outputs of at least 10^{12} neutrons/second. The work described leads to a specification for a neutron generator using the D-T reaction. One of the requirements of the generator is that the target area shall be of the order of 10 cm^2 and the overall size is limited by the need to surround it with a thick steel shield (50 cm).

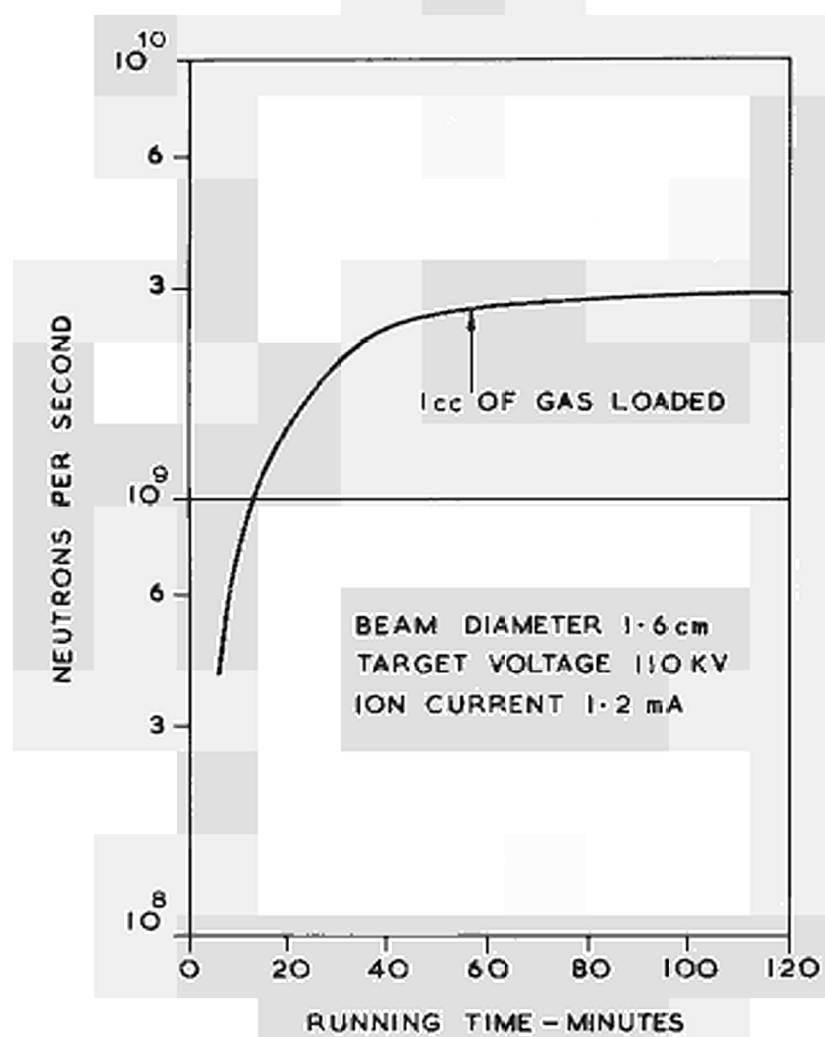


FIG 13. SELF LOADING OF MOLYBDENUM.

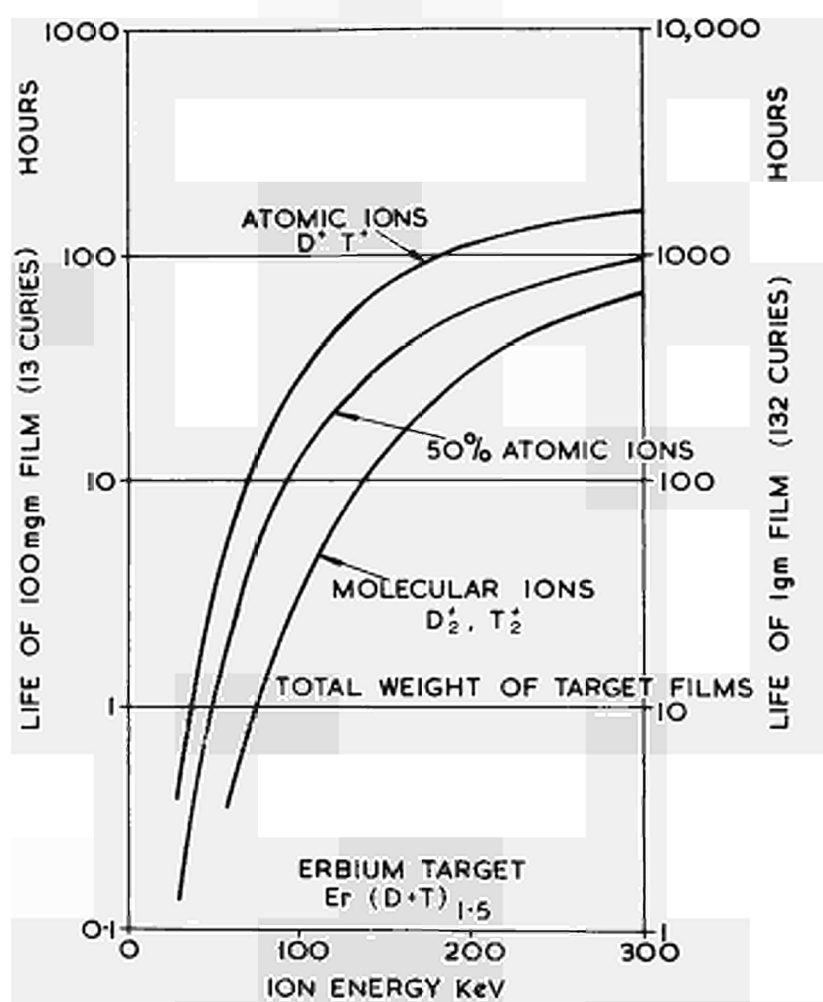


FIG 14 SPUTTERING LIFE OF TARGETS
AT 10^{12} NEUTRONS/SECOND

The dilution problem necessitates the use of a D-T mixture and as we have shown this leads to the use of voltages of 150 - 300 kV, since the neutron output/joule is a maximum at these levels and the cooling problem hence less severe. At 250 kV the ion current required is between 15 and 35 mA depending on the proportion of atomic ions. The life of the target is determined by sputtering. The sputtering life as a function of energy for two target thicknesses and three ion beams is shown in Fig. 14. This does not make any assumptions concerning dependence of sputtering coefficient on ion energy but merely takes into account the lower ion currents needed at higher voltages. As can be seen with thick targets it should be feasible to achieve target lives of several hundred hours at 10^{12} neutrons/second, provided the ion energies are over 200 keV. These figures are pessimistic in view of the expected decrease of sputtering with ion energy.

The clean conditions necessary for such a performance can only be achieved with a sealed accelerator of the type developed for 10^{10} neutrons/second and 10^{11} neutrons/second. A sealed system is also essential in view of the health hazards associated with the use of large amounts of tritium.

Acknowledgments

We acknowledge the valuable contribution of all those at S.E.R.L. concerned with the many aspects of tube construction, target evaporation and instrumentation.

Acknowledgment is also made to the Ministry of Defense for permission to publish this paper.

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DISCUSSION

Mr. SMITH

I also pointed out that the life I quoted was for static targets and not specifically for rotating targets.

Will you have a heat conduction problem with the very thick Erbium or Titanium Hydride targets ?

Mr. WOOD

Assuming the worst value for conductivity, calculation has shown no temperature in the target to rise above 600°C.

LONG-LIVED TARGETS

P. L. Jessen
Kaman Nuclear
Division of Kaman Corporation
Garden of the Gods Road
Colorado Springs, Colorado

ABSTRACT

Targets of exothermic materials such as zirconium and titanium have traditionally been most popular for low-energy accelerator neutron generators using the $T(d,n)He^4$ reaction. This is, of course, because of their high yield for given conditions.

The use of these materials in high power sealed-tube (non-pumped) accelerators presents several problems. These materials have low thermal conductivity when saturated with hydrogen isotopes. It is therefore necessary to use thin layers if the target material is to be cooled adequately to prevent outgassing. If thin layers (in the range of 1 - 5 microns thick) are used, however, they are sputtered away in a short time and the useful life of the target is ended.

Use of endothermic materials can eliminate this problem. Measurements of neutron yield from endothermic targets using molecular deuteron beams in the energy range of 130 - 200 kv show that these materials will provide higher yields than previous data would have suggested. This is especially true for ion beams consisting of a mixture of deuterium and tritium. Endothermic materials having good thermal conductivity (such as copper) can be used in relatively thick layers because the diffusion coefficient for hydrogen isotopes in many of these materials is quite low. If the ion beam is used to load the target material, the gas can therefore be concentrated near the bombardment surface of the target. It is thus possible to maintain the high thermal conductivity of the target layer throughout all of its thickness, except for the very thin layer near the bombardment

surface. A further advantage accrues from the use of a mixed ion beam (deuterium and tritium). The penetration of tritons will not be as great as that for deuterons at a given accelerating energy. Deuterons will therefore pass through the tritium-rich zone with greater energy than that with which they will pass through the deuterium-rich zone of the target. A higher neutron yield will therefore result than would be indicated by measurements using unmixed ion beams. The data indicate that approximately twice as much yield is obtained relative to that indicated for pure ion beams.

The results of measurements of neutron yield for some of the endothermic materials show that efficiencies of 30% relative to tritiated titanium are possible for copper among others. Those having high hydrogen solubility and relatively low atomic number are of interest.

The data show that sealed accelerators having yield of 10^{11} n/sec (4π) and good pressure stability can be constructed using these endothermic materials.

Operational simplicity and reliability of nonpumped or "sealed" miniature ion accelerators for neutron production have prompted the development of higher yield units of this type. Target materials for these accelerators must possess several significantly different properties than those for conventional Cockroft-Walton equipment. Conventional target materials which possess high thermal solubility for hydrogen isotopes do not fit these properties well in the following important respects:

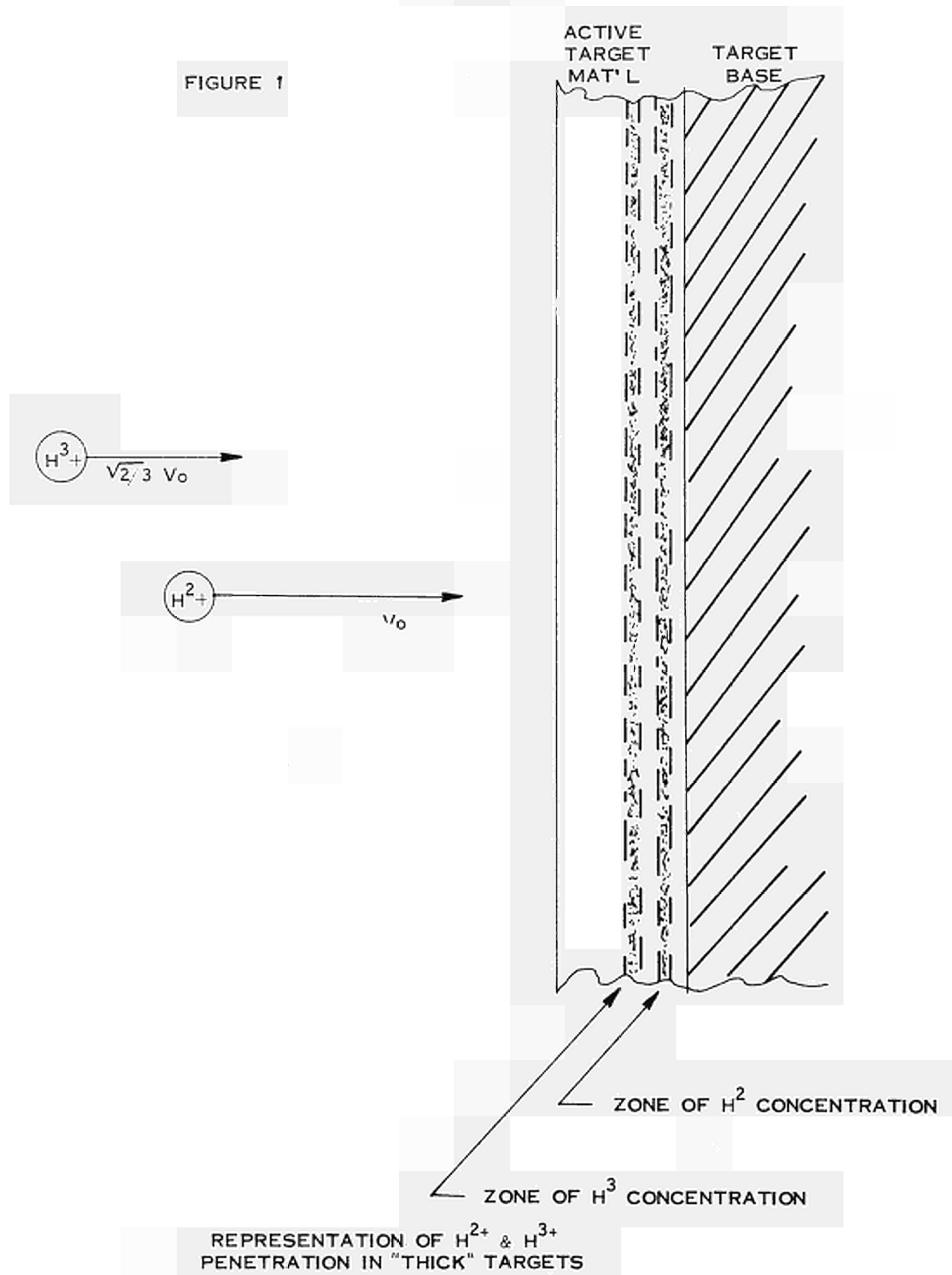
1. It is obviously not possible to frequently change the target in these accelerators if their advantages of economy and operational simplicity are to be maintained. This means that the sputter lifetime of the target layer should be significantly greater than that required for pumped Cockroft-Waltons. It is impractical to use extremely thick targets of the conventional metal hydrides. This is true because the hydrides of zirconium and titanium possess poor thermal conductivity. If a large amount of power is deposited on one surface of the target layer and if the opposite side of the target layer can practically be maintained at some given temperature, the poor thermal conductivity will result in a high temperature at the bombarded surface of the target. These hydrides are also exothermic, i.e., they give up occluded hydrogen as their temperature is increased because their hydrogen solubility is a decreasing function of temperature. Since these accelerators possess little pumping capability (and frequently none at all), this evolution of hydrogen isotopes creates a pressure stability problem.
2. It is necessary to operate these accelerators using a

mixed ionizing gas. This mixture usually consists of roughly equal parts of deuterium and tritium. If this ratio is perturbed significantly, the yield of the accelerator will decrease. Target materials which out-gas significantly will perturb this ratio. This occurs because the tritium will not penetrate the target layer to as great a depth as the deuterium for a given accelerating voltage. This is illustrated in Figure 1. If a thermal gradient exists across the target layer, as discussed in the previous section, the evolution of tritium will be more rapid than the evolution of deuterium, simply because the tritium will be concentrated in a zone of the target layer that is hotter than the zone in which the deuterium is concentrated.

3. If it were considered feasible to maintain the cooled surface of the target at a sufficiently low temperature to prevent outgassing of exothermic hydride targets, an additional difficulty still exists. The hydrogen diffusion coefficient of these exothermic metals (such as titanium and zirconium) is, of course, quite high. This was one of the criteria originally used in choosing these materials, since this is a necessary condition for efficient thermal loading of the metal with hydrogen isotopes. If a thick layer of these metals is used in order to obtain a good sputtering life, a large amount of tritium will be required because the tritium will diffuse in a short period of time throughout the entire layer. In other words, it is not possible to concentrate the hydrogen isotopes only in a thin layer near the bombardment surface. This means that the cost of the target will be appreciably greater, making the sealed accelerator less attractive from an economic standpoint.

It should be noted that these comments also apply (although

FIGURE 1



to a lesser degree) to other exothermic materials such as yttrium and erbium.

The difficulties of designing a suitable cooling system for sealed accelerators utilizing zirconium or titanium hydride targets may not be immediately apparent to the casual observer. Both calculation and experiment have indicated that if a 20-micron thick titanium hydride target is to be used in a sealed accelerator which deposits 400 W/cm^2 on the target surface (this would correspond to the order 10^{11} n/sec for a 200 kv molecular accelerator), the cooling system must be capable of maintaining the cooled surface of the target at a temperature of approximately -100°C . The cooling system must also permit close access to the target if an efficient target-to-sample geometry is to be maintained. To date the writer is not aware of any satisfactory designs for cooling systems that will meet these requirements, except for those in which the sample is placed in the cooling medium. This method of operation severely reduces the utility of the system.

These considerations make it apparent that other materials might serve as more suitable targets, even if the efficiency of these materials in terms of neutron yield per unit charge at a given accelerating energy was lower than that for titanium or zirconium. It may be seen that a desirable target material should have a reasonable solubility for deuterium and tritium, and that the solubility of hydrogen isotopes in the material should increase with temperature rather than decrease. Further, if the hydrogen isotope is placed in the material by bombardment, a large diffusion coefficient for hydrogen isotopes for thermal loading is not a necessity and is, in fact, undesirable. It was further felt that solubility, as measured by normal means, would not be particularly significant if the hydrogen isotopes are accelerated into the material. Previous investigations of "drive-

in" targets by Reifenschweiler and others¹ indicated that more effective drive-in targets could be made than would be indicated by comparing the thermal solubility of these materials (such as gold) to more conventional ones (such as titanium and zirconium). It was felt, however, that the amount of hydrogen that could be injected in the material by ion bombardment might well be proportional to the thermal solubility of hydrogen in the material. Copper, manganese and nickel were investigated because these metals all exhibited an increasing solubility for hydrogen with increasing temperature. The materials are available in reasonably pure forms that are suitable for target fabrication and their diffusion coefficients for hydrogen are relatively low at room temperature. Figure 2 shows the variation of hydrogen solubility with temperature for copper, manganese, nickel and titanium. It may be seen that the thermal solubility for titanium is much higher than that for the other three materials.

Table I shows the relative efficiencies of the four target materials following a sufficiently long period of operation that the neutron yield was stable with time. Again, it may be seen that the efficiencies of all these materials are surprisingly high relative to titanium in view of the low thermal hydrogen solubilities of these materials with respect to titanium. This unexpectedly high yield implies that the target material is retaining a much greater amount of the ionizing gases than thermal solubility data would suggest. It further suggests that the gas must remain concentrated in a thin volume near the surface of the target. The range of deuterium and tritium ions in these materials is of the order of 0.5 microns. The variation of this range for

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FIGURE 2

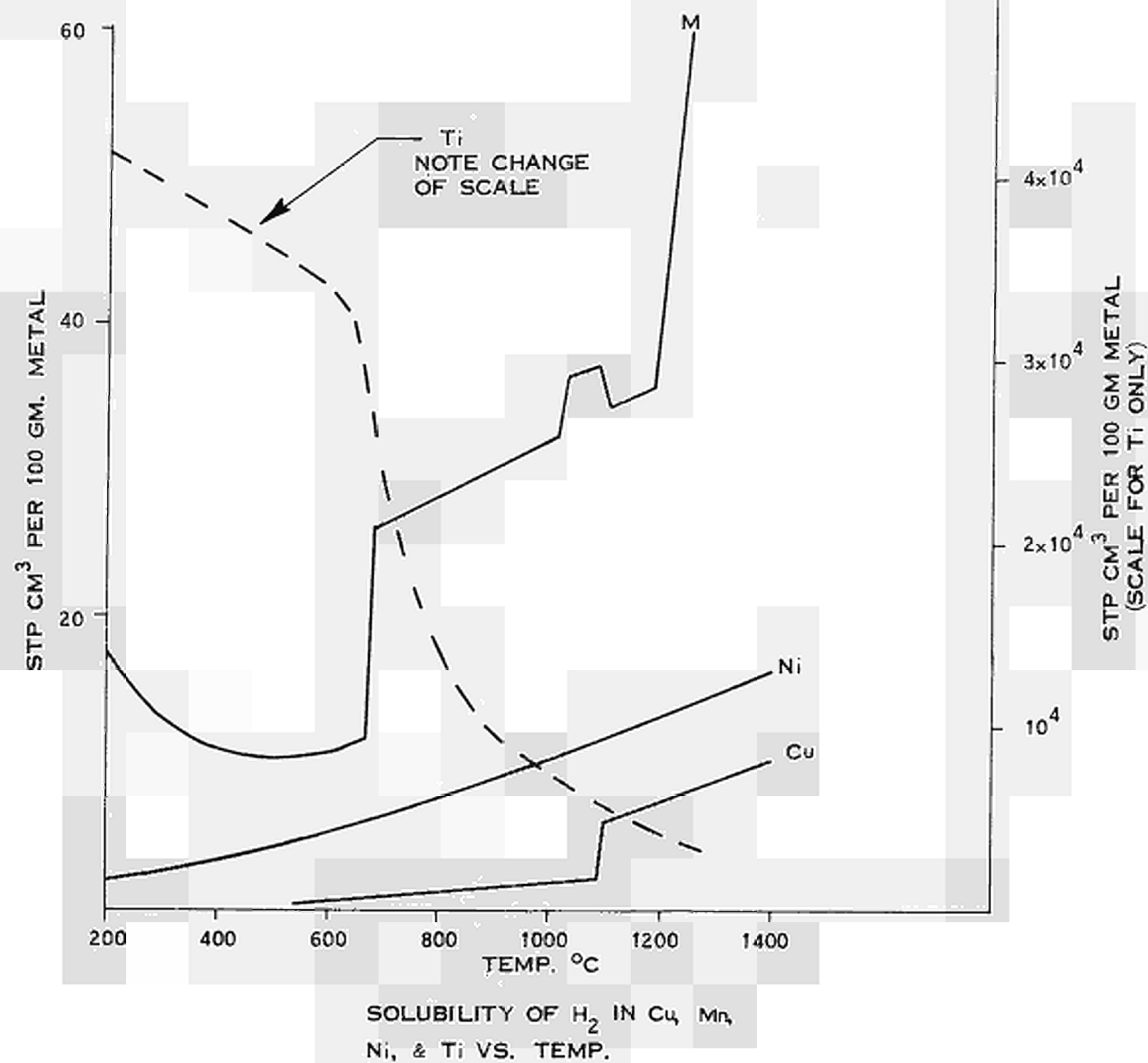


TABLE I

Comparison of "drive-in" target materials to a TiD target of 1:1 Ti to D ratio, "drive-in" targets bombarded at 130 kv, yield measured at 160 kv.

<u>Material</u>	<u>Yield Relative to TiD Target</u>
TiD	100%
Ni	50%
Mn	42%
Cu	22%

different ions, or "straggling," is less than 10% of the range. It would therefore be expected that the gas would be initially deposited in a fairly small and well-defined zone. If the gas is to move away from this zone, the mechanism for this movement would be either diffusion or collisions from other energetic ions.

The low diffusion coefficient of these materials is undoubtedly important in maintaining a high concentration of gas in a zone in which impinging ions have sufficient energy to permit significant neutron production.

Collisions are not significant to the gas movement problem, since collisions between the impinging ions and the trapped gas atoms in the target are precisely the necessary condition for neutron production.

The targets were evaluated by two different methods. Initial evaluation of the copper targets was performed by mounting a copper foil on which a film of titanium having a thickness of approximately 1.5 microns had been evaporated in a sealed accelerator. This accelerator was then operated using a mixture of deuterium and tritium as the ionizing gas for a period of time much longer than that required to completely sputter away the titanium layer. This permitted a direct comparison of titanium and copper targets, while providing the least chance that changes in the accelerating system would distort the comparison data. A summary of runs for three different sealed accelerators is shown in Figure 3. These sealed accelerators were started in operation using pure deuterium as the ionizing gas with all of the tritium in the tube being occluded in the titanium layer on the target. As the tube operates, deuterium is driven into the target and tritium is evolved. The neutron yield of the accelerator for given voltage and current conditions decreases as an equilibrium gas mixture is established. The ultimate equilibrium ratio will be determined by the total amount of deuterium and tritium

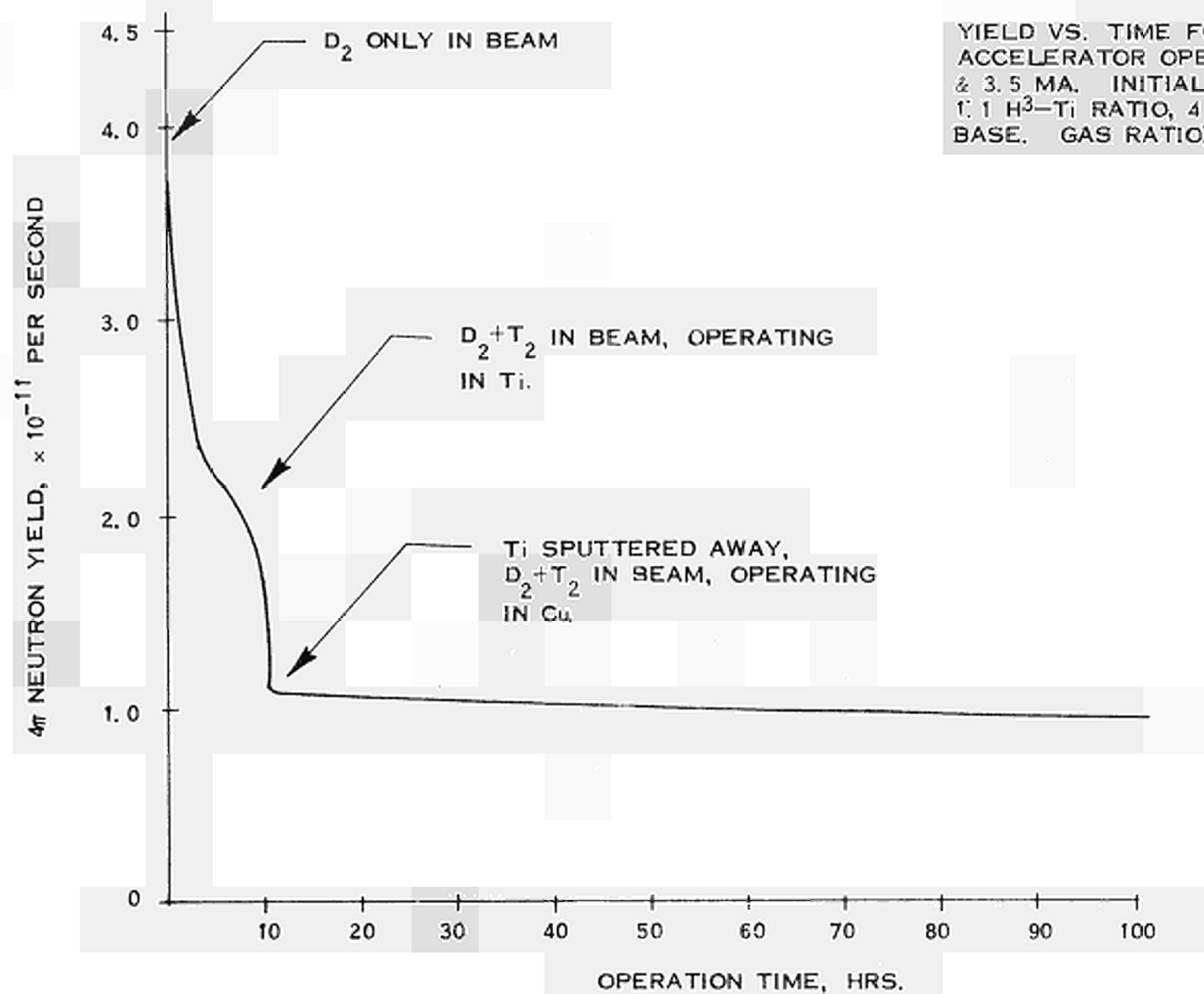


FIGURE 3

YIELD VS. TIME FOR TYPICAL SEALED ACCELERATOR OPERATING AT 190 KV & 3.5 MA. INITIAL TARGET CONDITION: 1:1 H³-Ti RATIO, 4C/IN², ON COPPER BASE. GAS RATIO: \approx 1:1.

present in the tube, together with the effect of differential evolution rate of the two gases from the target, as previously discussed. The neutron yield will then decrease further as the titanium layer on the target is sputtered away. A new equilibrium value will then be established when the titanium layer is essentially totally removed. This condition occurs in approximately 20 hours of operation for the particular accelerator in question. Following this event the neutron yield remains relatively constant for the rest of the test interval. It may be seen that the copper is a surprisingly effective target material, relative to titanium, in view of the fact that the hydrogen solubility in titanium is roughly three orders of magnitude greater than that for copper.

This method of evaluation was discontinued because of the complexity and expense involved. The remainder of the target evaluations were performed utilizing a conventional Cockroft-Walton accelerator in which targets could be readily changed. The vacuum in the accelerator was quite clean. A sputter ion pump was used, and the critical accelerator seals were all metallic. This reduced target surface contamination effects that plagued several earlier investigations. It should be mentioned that all of the accelerators used in this work utilized a Penning ion source that provided an unanalyzed ion beam which was approximately 93% molecular. The Cockroft-Walton accelerator was not operated in a mixed beam mode, but was instead operated on pure deuterium. The efficiency of the targets was compared to both copper and to saturated titanium deuteride targets so that the data could be related to that obtained for the sealed accelerator. The targets were loaded with deuterium by operating the accelerator at a voltage equal to the square root of $\frac{2}{3}$ times the voltage at which the yield was measured in order to simulate the lesser penetration of the tritium ions which would occur in the mixed beam operating mode. Results comparable to those for the sealed accelerator with a copper target were obtained by this means.

Targets were prepared by different means, depending upon the characteristics of the particular material. The copper targets were prepared simply by punching 1-1/2-inch discs from 0.010-inch thick OHFC copper, which is available as a commercial product. The manganese targets were prepared by evaporating a 5-micron layer of manganese on a copper substrate that was essentially identical to the copper targets. The manganese used for the evaporation was stated as 99.97% purity. The evaporation was performed at pressures of less than 10^{-6} mm of mercury to maintain the purity of the evaporated film at a reasonable level. The nickel targets were formed by bonding a 0.001-inch thick nickel foil of 99.97% purity to a copper substrate using 6040 tin-lead solder. The titanium targets were prepared by evaporating a 1.5-micron thick film of titanium on a copper substrate, which again was identical to the copper target. These targets were prepared as a commercial product by the U. S. Radium Corporation of Morristown, New Jersey, U.S.A.

Prolonged periods of operation and prolonged periods of storage of accelerators utilizing copper targets provide yield data as shown in Figure 4. These data indicate that the diffusion coefficient in copper is sufficiently low that no significant migration of the hydrogen isotopes in the copper occurs. This would indicate that the position of the gas in the target is relatively stable (which would be suggested by the low diffusion coefficient for copper) and would further indicate that accelerators using these targets can be stored for considerable periods with little deterioration of their operating characteristics.

The data indicate that targets having useable efficiency can be obtained by bombarding these materials with a mixture of deuterium and tritium ions. The operational data for the targets in sealed accelerators indicate that they do provide significant advantages in terms of lifetime and pressure stability. At first

FIGURE 4
YIELD OF A SEALED ACCELERATOR
AT VARIOUS TIMES OF CONTINUOUS
WEEK OPERATION. (NOT IN CONTINUOUS OPERATION)

RELATIVE YIELD

1.0
0.8
0.6
0.4
0.2
0

0

1

2

3

4

5

6

TIME IN WEEKS

RELATIVE YIELD

1.0
0.8
0.6
0.4
0.2
0

0

1

2

3

4

5

6

TIME IN WEEKS

glance the neutron production efficiency appears to be relatively poor, i.e., of the order of 20 - 40% for the various materials. As an example, the sealed accelerator in which these targets were operated is capable of providing 3.5 milliamperes of molecular ions at 200 kv total accelerating potential. These operating conditions using a pure deuterium beam on a tritiated titanium target having a 1:1 atomic ratio of tritium to titanium would yield approximately 4.5×10^{11} n/sec. Using a pure copper target, a yield of approximately 1×10^{11} n/sec was obtained. This represents an efficiency of only 22% relative to a conventional Cockroft-Walton accelerator operating with deuterium as the ionizing gas and tritium in the target. If one considers the application of these accelerators to activation analysis, the efficiency looks considerably different, however. In activation analysis applications, there will be some minimum yield which is considered to be useable because of the relationship between yield and the precision of the analysis. A rather large group of analyses require yields of the order of 5×10^{10} n/sec (4π). A conventional Cockroft-Walton accelerator operating with a pure deuterium beam and having a maximum rated yield of 2.5×10^{11} n/sec (this class of accelerator is quite common in the United States) can provide a yield of 5×10^{10} n/sec for approximately 10 hours if the target current is carefully adjusted upward as the target efficiency deteriorates. This also assumes that the vacuum system for the accelerator is an extremely clean one. This is necessary so that the level of heavy ion contamination in the beam is small. If this contamination level is not small, the rate at which the target is sputtered increases markedly and the target may deteriorate due to sputtering before the 10-hour lifetime can be obtained. In contrast, sealed accelerators operating with a mixed beam using copper targets have been operated in excess of 100 hours at levels in excess of 5×10^{10} n/sec, i.e., at a level of 10^{11} n/sec. The true lifetime of these sealed accelerators is not in fact known since the life tests were

terminated for other reasons. It is possible to conclude, however, that the use of these techniques will provide at least an order of magnitude improvement in the irradiation time that can be obtained from the neutron generating system before maintenance is necessary. A direct comparison of the economics of the two systems is not possible because of the wide variation in maintenance costs for different facilities. Some facilities in the United States have estimated, however, that the cost of changing a target in the typical Cockcroft-Walton accelerator, including the cost of the target, the labor involved, and the down time of the facility, lies in the range of \$50 - \$200 per target. The cost of ten target changes at this level is comparable to the cost of the regeneration of a sealed accelerator. This would make the use of sealed accelerators with low efficiency targets appear economically attractive, particularly in those facilities where labor costs are high and/or the tritium contamination associated with changing targets in Cockcroft-Walton accelerators creates significant decontamination problems or where such contamination is not permissible.

DISCUSSION

Mr. SMITH

When you produced a self loaded target, how long did it take to reach maximum output.

Mr. JESSEN

It was found to be quite variable depending on the material. At currents in the region of 1.0 to 1.5 ma, it ranges from 1/2 hrs for copper to 2 1/2 hrs for nickel.

Mme BREYNAT

Quelles sont les températures maxima des cibles de Mn ou Ni et le rendement neutronique de ces cibles ?

Mr. JESSEN

The maximum temperature at which we operated was 600°C, since our primary object was to alleviate the cooling problem, rather than high energy operation per se.

A DESIGN STUDY FOR A GAS TARGET USING A ROTATING FOIL *

Frank S. Williamson, Division of Biological and Medical Research,
Argonne National Laboratory, Argonne, Illinois, U.S.A.

SUMMARY:

There is a need for targets capable of accepting 1 milliampere of deuterons, at 3 MeV, for the production of monoenergetic neutrons. It is often an advantage for the neutron source area to be as small as possible even though a relatively large area is required for effective cooling. The greatest neutron output from the $D(d,n)^3\text{He}$ reaction is obtained from a gas target.

This study examines the feasibility of a rotating gas target. The deuteron beam enters a target chamber, containing deuterium gas at atmospheric pressure, through an annular nickel foil 10 cm in diameter and 0.5 cm wide. The foil is 1.27×10^{-4} cm thick. The target chamber, with the foil, rotates at 6000 r.p.m.

The deuterium, which is an excellent cooling gas, enters and leaves the target chamber through concentric tubes which are coaxial with the annular foil. The gas is circulated by an external blower and is cooled by a conventional gas/water heat exchanger in a closed loop. The gas is constrained to flow across the foil radially, outward with a linear velocity of 50 meters/sec., then over cooling fins inside the target chamber casing and again radially inward to cool the part of this casing which absorbs the residual energy of the beam. Temperature rise in the gas from dissipation of the beam energy would be about 80°C. Calculated foil temperatures would vary, in the rotation cycle, from about 250°C to 450°C when only convection losses due to turbulent gas flow are considered. Additional heat losses, due to radiation and conduction, in the foil can be expected to reduce these temperatures somewhat.

INTRODUCTION

For some years radiobiologists have appreciated that experiments on living organisms, utilizing monoenergetic neutrons, would yield information of fundamental importance. It is not difficult to obtain 14 MeV neutrons from the (d,t) reaction and many investigations have been carried out in this way. Neutrons of lower energies are more interesting, however, because they are relatively more effective in producing biological damage. Up to the present time few such experiments have been made on biological materials because of the low neutron dose rates which can be obtained using existing accelerators and targets.

A new generation of accelerators is now emerging which will be capable of producing at least 1 milliamperes of analyzed protons, or deuterons, at energies up to 3 MeV. This type of accelerator can, by utilizing the target reactions (p, ^7Li), (p,T), or (d,D), provide neutrons of any energy from a few keV to 6 MeV.

Attempts to increase the beam-current capacity of such targets have usually involved a large increase in the bombarded area either by defocussing or scanning the beam. Such a method is useful in biological irradiations under certain circumstances (Frigerio and Jordan).⁽¹⁾ In general, however, a target for use in radiobiology should have the smallest possible source area. It is well known that, in work with monoenergetic neutrons where only a small energy-spread can be tolerated, a thin target must be used and the angle subtended at the target by the specimen must be small. It is obvious that the angle subtended at the specimen by the target must also be small. Furthermore, even with the increased dose rates to be expected using higher beam currents,

* Work supported by the U. S. Atomic Energy Commission.

it will often be necessary to take advantage of the inverse-square law and obtain the maximum dose rate, which is available close to the target. The inverse-square law only applies when the target can be regarded as small, in comparison with the distance separating the target and the biological specimen.

This paper examines the feasibility of a gas target, for the $D(d,n)^3\text{He}$ reaction, which will accept a deuteron beam of 1mA at 3 MeV, and employ a very small bombarded area.

The $D(d,n)$ Target

The principle of the rotating target is well known. In this design it has been applied to a gas target. A beam of deuterons passes into a target volume of deuterium gas through an annular window-foil which is rotating at high speed. A second rotating element stops the beam after its passage through the gas. Both foil and beam absorber are cooled by a stream of the target gas flowing at a high velocity. The gas enters and leaves the target chamber through concentric tubes and is circulated through a heat exchanger by means of an external blower.

The arrangement is shown in Fig. 1 as a cross-section. Figure 2 is a cut-away drawing showing the view from one end of the assembly, in the direction of arrow 'A' (Fig. 1). In these figures constructional details have been omitted in the interests of clarity.

With the dimensions selected it is not practicable for the axis of rotation and the particle beam axis to be both co-planar and parallel. The two axes lie in parallel planes 5 cm. apart. The projection of the beam axis upon the plane containing the axis of rotation intersects this axis at 30° . This geometrical relationship is illustrated in Fig. 3.

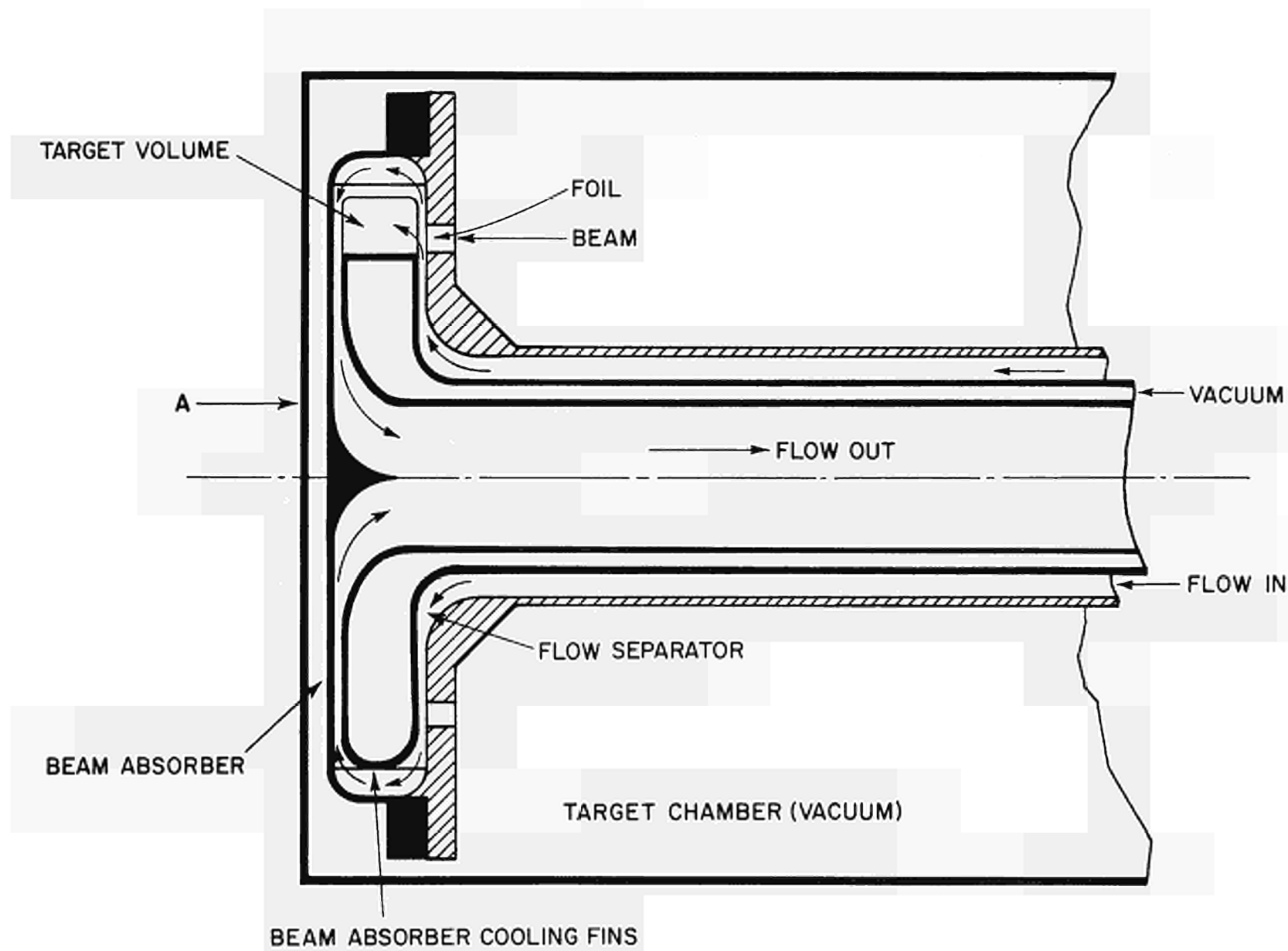


FIGURE 1

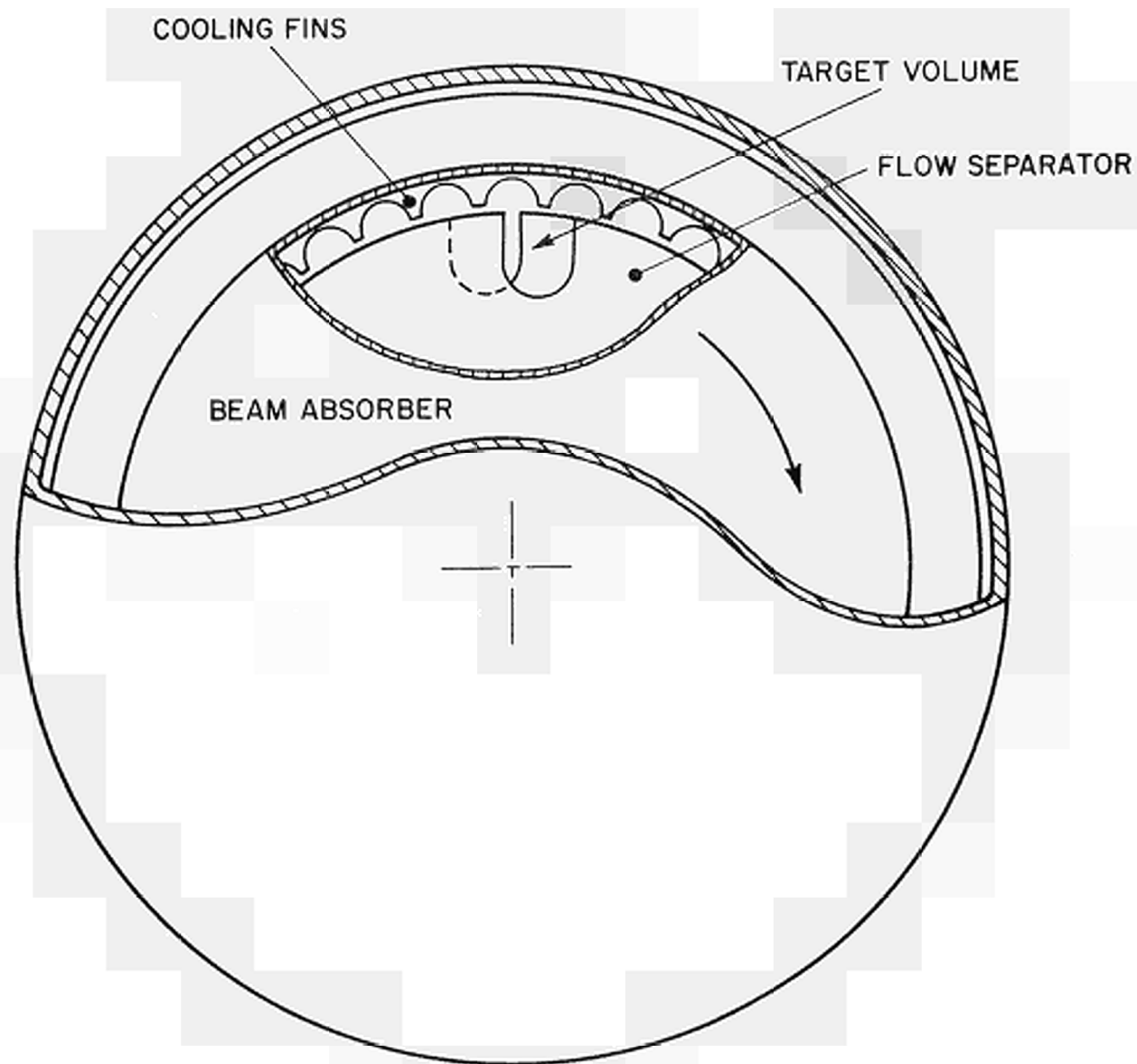


FIGURE 2

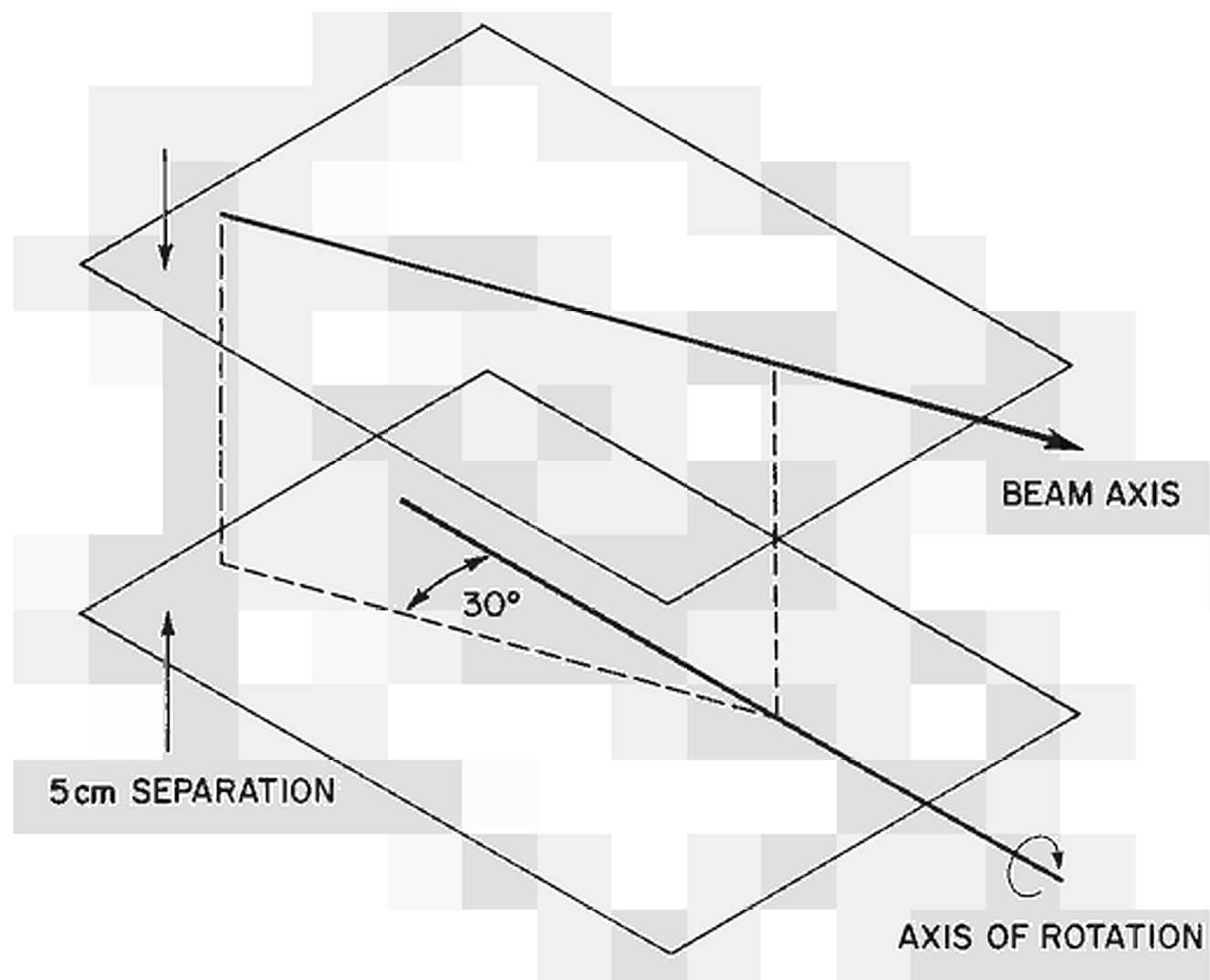


FIGURE 3

The Foil; Heating and Cooling

The first parameter to be fixed is the rotational speed of the target. This can readily be calculated from the rate of temperature rise of the foil, assuming no heat losses, during the brief period, t , when an element of the foil is under the beam.

If an element of the foil remains under the beam for t secs, then the temperature rise, ΔT_f is given by

$$\begin{aligned}\Delta T_f &= \frac{(\text{energy lost by beam}) \times (\text{beam current}) \times t}{(4.2 \text{ Joules/cal.}) \times (\text{beam area}) \times (\text{mass/cm}^2) \times (\text{specific heat})} \\ &= \frac{\Delta E \times I}{4.2} \times \frac{4}{\pi d^2} \times \frac{t}{m \times 10^{-3} \times c_f}\end{aligned}$$

t is the time taken for the element to pass through the beam.

$$\text{Hence (rotational speed)} = \frac{d}{\pi \times D \times t}$$

so that:-

$$\text{Rotational speed (rev/sec)} = \frac{96.5 \times I}{d \times D} \times \frac{\Delta E}{m \times c_f \times \Delta T_f} \quad \text{Equ. (1)}$$

where I = beam current (mA)

ΔE = beam energy loss in the foil (keV)

d = beam spot diameter (cm)

m = foil thickness (mg cm^{-2})

ΔT_f = allowable cyclic temperature rise in the foil ($^{\circ}\text{C}$)

c_f = specific heat of the foil material (cals/g)

D = mean diameter of the foil annulus (cm)

The quantity $\frac{\Delta E}{m \times c_f \times \Delta T_f}$ is a useful guide for material section.

Using data for ΔE and m given by Coon⁽²⁾ for the foils commonly used in gas targets, and assigning arbitrary values of ΔT_f , we obtain for aluminum ($\Delta T_f = 100^{\circ}\text{C}$), a value of 7.84. Similarly nickel ($\Delta T_f = 200^{\circ}\text{C}$) gives 5.16 and molybdenum ($\Delta T_f = 400^{\circ}\text{C}$) gives 4.26.

The material yielding the smallest value of this guide number would be preferred. While these numbers are only based on estimates of ΔT_f , there can be no doubt that either nickel or molybdenum would be superior to aluminum. Since molybdenum is only available about 1 MeV thick for deuterons, nickel becomes the material of choice in this design.

The rotational speed required is not influenced by the foil thickness since $\Delta E/m$ is approximately constant for a particular material. The total heat absorbed in the foil will, however, be proportional to thickness, and this must be small enough to be dissipated by the cooling gas stream.

For the chosen parameters: -

$$\begin{array}{ll} I = 1 \text{ mA} & \Delta T_f = 200^\circ \text{ C} \\ \Delta E = 140 \text{ keV} & c_f = 0.12 \\ d = 0.5 \text{ cm} & D = 10 \text{ cm} \\ m = 1.13 \text{ mgm/cm}^2 \end{array}$$

Rotational speed = 99.6 rev/sec or approximately 6,000 rev/min. The foil may be mounted on the foil carrier by soldering. The annular gap through which the beam passes is not complete, but has three radial webs to support the outer ring. The webs are inclined at 30° to the plane of the foil carrier so as to present the minimum obstruction to the inclined beam.

The deuterium gas flows radially over the surface of the annular foil. The calculation of heat loss to the gas can only be approximated. If the gas stream is confined to a region 0.2 cm thick, where it flows over the foil, the problem becomes analogous to that of a gas flowing between concentric tubes.

In such a case the equivalent diameter (D_e) of a single tube is defined as $D_e = 4 A/C$, where A is the cross-sectional area of the annulus

and C its circumference.⁽³⁾ In this case $D_e = 0.8$ cm. The average heat transfer coefficient for turbulent flow is given by h in the dimensionless equation: -

$$\text{Nusselt's Number } N_{Nu} = \frac{hD_e}{k} = 0.023 (N_{Re})^{0.8} (N_{Pr})^{0.4}$$

$$N_{Pr}, \text{ Prandtl's Number,} = 0.71 \text{ for gases}$$

$$N_{Re}, \text{ Reynolds Number,} = \frac{vD_e \rho}{\mu}$$

For Deuterium at standard temperature and pressure, in MKS units;

$\rho = 0.017 \text{ kg m}^{-3}$ and μ (Dynamic viscosity) is estimated to be

$$0.88 \times 10^{-5} \text{ kg m}^{-1} \text{ s}^{-1}$$

$$D_e = 8 \times 10^{-3} \text{ m}$$

The estimated thermal conductivity for Deuterium, $k = 15 \times 10^{-2} \text{ J m}^{-1} \text{ s}^{-1} \text{ deg. C}^{-1}$

$$\text{Hence } h = 320 \text{ J m}^{-1} \text{ s}^{-1} \text{ deg. C}^{-1}$$

$$N_{Re} = 4595$$

For a Reynolds number larger than 2300 the flow will be turbulent, as required.

It is now necessary to include the effect on $(\Delta E \times I)$ of the 30° angle of beam incidence.

$$\text{Hence energy absorbed} = \frac{140 \text{ keV}}{\cos 30^\circ} \times 1 \text{ mA} = 162 \text{ J s}^{-1}$$

$$\text{The foil area} = 1.57 \times 10^{-3} \text{ m}^2$$

Consequently the mean foil temperature is 322° C above that of the cooling gas, or approximately 350° C .

During rotation the foil temperature will cycle between approximately 250° C and 450° C .

The effective gas velocity across the foil will be increased in some measure by the rotation of the target. The peripheral velocity is 31.4 m s^{-1} . It seems reasonable to assume that the gas will acquire some of this velocity

component, but probably not more than 50% of it. The resultant gas velocity relative to the foil should be at least $(50^2 + 15.7^2)^{1/2}$; approximately 52.4 m/s. Since the heat transfer coefficient h is proportional to $(\text{velocity})^{0.8}$, it is apparent that this additional velocity component will have an insignificant effect.

Both radiation and conduction mechanisms will remove some heat from the foil. At the peak cyclic temperature of 450°C the rate of cooling due to radiation will be 15% of the rate due to forced convection. At the mean temperature, 322°C , the radiation cooling contribution is 11% and at the minimum temperature, 250°C , it falls to 8%.

The calculation of heat loss by conduction radially along the foil to the foil carrier is obviously complex. For example, when an element of the foil has just emerged from under the beam, the heated area extends right up to the edge of the foil where it is cooled by its support. This boundary region must lose heat almost instantaneously. In order to make a steady-state approximation, the uniformly heated zone in the annulus is taken as extending to within 1 mm of the support, and the conduction loss calculated for a nickel foil 1.27×10^{-4} cm thick, of total periphery 63 cms (inner and outer combined). At a mean temperature difference of 300°C over the 1 mm length, the energy lost will be about 3.2 cal. s^{-1} , or 13.4 J s^{-1} . The energy input is 162 J s^{-1} so the loss due to conduction is approximately 8%.

The combined heat loss due to radiation and conduction will make a useful contribution to the foil cooling.

At these foil temperatures there will be some diffusion of deuterium. This gas would be removed by the Vac-Ion pump which should, in any case, be installed on the accelerator side of the target for the purpose of minimizing contamination of the foil.

The Beam Absorber

After the beam has lost 162 keV of energy in the foil, followed by 104 keV in the gas target volume, it will still have up to 2.73 MeV energy remaining. This energy is completely absorbed in the rotating casing which also contains the gas system. Since the disc is rotating at the same speed as the foil, and the beam area may be taken as unchanged, the required thickness for an acceptable cyclic temperature rise of $\Delta T_a = 100^\circ \text{ C}$ may be calculated using Equation (1)

Taking copper as a possible material m must be approximately 60 mgm/cm^2 .

Since the thickness of the beam absorber is not likely to be less than 1 mm, ΔT_a will be less than 9° C .

The beam power dissipated in the absorber has to be removed almost entirely by the cooling gas. For this reason the gas flow is forced into a region 0.2 cm thick as in the case of the foil cooling. The inside of the periphery of the absorber is equipped with axial fins to increase the surface area exposed to the gas stream.

The gas velocity in the layer bounded by the foil is 50 m s^{-1} . This corresponds to a flow of 31.4 liters/sec. If all the 3 kW of beam power is eventually dissipated by the gas, its temperature will increase by approximately 80° C .

The Gas Flow System

The oblique beam path through the gas corresponds to an energy loss of 104 keV at S.T.P. for the 2 cm of gas 'thickness' allowed in this design.

In order to maintain high velocities in the gas for effective cooling it is necessary to include the shaped flow separator (Fig. 1). This is

combined with a double-walled tube which separates the entry and exit flow streams. It is convenient to construct this so that the flow separator and tube is stationary, so that only two rotating seals are required (one for the entry flow tube and one for the vacuum system).

The flow separator is cut away to allow passage of the beam. This cavity, referred to in Fig. 1 as the Target Volume, is continuously purged by deuterium from the revolving fins on the beam absorber, and maintained in an extremely turbulent state. This provision should minimize spurious density effects in the gas produced by beam heating.

The gas velocity in the exit tube is 57.4 m s^{-1} , which results in a Reynolds Number of 15,360. This will certainly mean a turbulent flow condition, which is unavoidable, but the pressure drop can be shown to be negligible.

In order to prevent the entry and exit tubes from functioning as a contraflow heat exchanger, the double-wall of the tube, and the flow separator, are maintained under vacuum.

Conclusion

This study indicates that a rotating-foil gas target capable of handling a beam current of 1 mA is feasible.

The foregoing calculations have all been made using steady-state solutions to the heat transfer problem. These tend to provide an upper limit to estimates of temperature. Before constructing such a target it would be helpful to perform a more detailed heat transfer analysis based on non-steady state treatments.

For the present we can only speculate on the effects on the foil of a high velocity turbulent gas flow coupled with repeated, rapid, temperature cycling. Such questions can best be resolved by experiment.

If a prototype is constructed, it would be a good idea to test it using a deuteron beam of approximately 160 keV energy. In this way the foil system could be fully tested, using deuterium gas, without any concern about the cooling of the beam absorber, and without producing a dangerous neutron background. By increasing the energy of the deuterons to approximately 300 keV, the effectiveness of the gas circulation in the target volume could also be checked. Such testing could be carried out before a 3 MeV, 1 mA deuteron accelerator is available.

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DISCUSSION

Mr. RICCI

Have you calculated what neutron output you may expect with this target ?

Mr. WILLIAMSON

This has not been calculated because the requirement is simply to obtain the maximum possible output. But if we roughly assume that 500 μ A of 1 MeV deuterons on a thick D₂O ice target would produce about 10^{10} neutrons per second, then this thinner gas target might be expected to produce about the same output.

Mr. PAIC

What is the beam spot size ?

Mr. WILLIAMSON

The width of the annular foil was chosen to be 5 mm because the available data on foil performance applies to a foil of 3/16 inch diameter. The beam spot just fills this 5 mm diameter. I would like to point out that further relief of the cooling problem would be obtained by resorting to a square beam spot, rather than circular.

A rotating heavy ice target for use in the production
of fast protons by the He^3 (d, p) He^4 reaction

by

D.A. Bance

Medical Research Council, Radiobiological Research Unit,
Harwell, Didcot, Berkshire, England.

Summary

The development and performance is described of a heavy ice target for use with nuclear reactions involving the bombardment of deuterium. Evaporation of the heavy ice under bombardment is minimised:-

- a) by cooling with liquid nitrogen
- b) by having the ice layer on an annulus of mean diameter 3 inches and width 1.75 in., which rotates continuously under the beam at 1000 r.p.m.
- c) by scanning the beam at 100 Kc/sec. so that the effective area is approximately 1.5 sq. inches.

The target has been used for the He^3 (d, p) He^4 reaction for production of 14.5 MeV protons for use in radiobiological research. Stable outputs have been obtained at 150 watts on target for several hours. The proton emission strength was 9×10^9 protons/sec. at this power.

1. Introduction

In the research programme at the Radiobiological Research Unit at Harwell a source of mono-energetic protons to investigate the biological effects of ionizing radiation was desirable. To fill this need it was decided to adapt a 1 MeV Cockcroft-Walton accelerator, which up to this point had been in use for the production of neutrons (Neary, 1964). This accelerator, which had been designed and manufactured by Philips of Eindhoven, was very suitable for this purpose, since the target assembly was particularly easy to modify and already had many desirable facilities incorporated, such as beam alignment, beam scan, etc. The RF ion source was also readily adaptable to take a new gas control system.

The nuclear reaction considered most suitable for this work was the Helium 3 - Deuterium reaction. This could be effected either by the acceleration of deuterium ions into a Helium 3 gas target, or by the acceleration of Helium 3 ions onto a solid target containing deuterium. The latter method was chosen as that presenting the fewer technical problems and certainly the least expensive in capital equipment costs.

For a relatively quick and easy method to prove the possibilities of such a system and its initial application, a deuterium-enriched Li^6 target similar to those we had used successfully for neutron production, (F.S. Williamson, 1964) was placed in the beam. This was found to give satisfactory results but it was soon found, however, that the dose rate from such a target was insufficient for experiments requiring high dose rates and that the life of the targets was limited to a few hours. To improve this latter limitation substitution of liquid nitrogen cooling for the original water cooling was made. This gave longer target life but the necessity for a higher dose rate remained a problem. Various other targets were used such as titanium deuteride and natural-lithium deuteride but with no significant improvement in output although these

targets were used successfully for many biological experiments. (Neary and Savage 1966; Neary 1967; Munson, Neary, Bridges and Preston, in press). An experimental study of the use of stationary D_2O targets, cooled by pumped liquid N_2 , was carried out. A three fold increase in output, compared with the previous best Ti D. target, was obtained but for only relatively short periods. The maximum heat input permitting stable running was 60 watts (600 KV, 100 μA). Rapid deterioration occurred if 100 watts was dissipated. Output fell by 60% over a period of 90 mins. Beam scan was employed for all experimental runs, the target area being 1 sq inch.

It was at this stage that the decision to develop a liquid nitrogen cooled rotating heavy-ice target was taken.

2. General design features.

The problem involved were not unknown to us, as some 12 years previously a similar target had been developed and supplied by Philips of Eindhoven, for the production of neutrons. Details of the design of this target have been published (J.H. Spaa, 1957). There were certain aspects of the design of this equipment that made it unsuitable for our particular application and so it was decided to start afresh. The basic requirements thought necessary in the design were:-

1. the rotating target should be a hollow Cu container, the inside of which was to serve as the liquid nitrogen reservoir with the facility for easy replenishment as boil-off occurred. The outside of the container was to be shaped so as to have an annular surface which would be the target face, this face to be at an angle of 97° to the incident beam of Helium 3 ions.
(Note. At the angle of 97° the energy of the resultant proton is independent of the energy of the Helium 3 ion, the proton energy being 14.5 MeV).
2. the rotational speed of the target should be approximately 1000 rpm and the mean diameter of the target annulus should be 8 inches. This would give a target surface speed of 420 inches/sec.
3. the rotary vacuum seal should be able to maintain a working vacuum pressure for at least 50 hours of rotation; the replacement of the vacuum seal should be a simple operation.

4. a simple but accurate method of D_2O dispensing should be aimed at. The usual method of glass pipettes, ground glass joints and grease-laden taps was to be avoided.

Certain other requirements such as a beam stop flap, a target tube cold trap and a secondary emission suppression facility were already available in the target tube as was beam scan equipment. At this stage the firm of Multivolt Ltd., was approached, and agreed to undertake the manufacture of the main parts of the equipment.

3. Details of design.

Figure (1) shows the basic design of the target dish. The liquid nitrogen filler tube is concentric with an outer sleeve which acts as the drive and bearing shaft and also, at its lower end, the sealing surface for the rotary vacuum seal. A high carbon - manganese steel was used for this sleeve to enable the sealing surface to be hardened and finely ground, (less than 8 micro inch surface roughness). This steel also has low thermal conductivity properties.

A synthetic rubber "Lip seal" was chosen for the rotating shaft vacuum seal, its advantage being the small contact area and hence, minimum frictional resistance and heat rise. It will be appreciated that under the stringent conditions required in this application, such a seal must be run with negligible axial 'throw' on the shaft. To meet this requirement the bearings used were of high precision, and the lower race was placed as close as possible to the rotary seal. The target assembly was also dynamically balanced.

Figure 2 shows the method used to introduce the liquid N_2 into the rotating container. Coolant from a pressurised dewar vessel is fed down the small diameter centre tube. The lower end of this tube has an increased diameter and slots in this section mate with holes in the base of the target tube. This configuration facilitates introduction of the coolant when the target is rotating and also allows a free passage to atmosphere for the exhaust gas.

It was felt that the dispensation of heavy water should be a simple and accurate operation. The system used is shown in Figure 3, a hypodermic syringe being used to transfer a known amount of liquid through a "Subaseal"

ROTATING D₂O ICE TARGET .

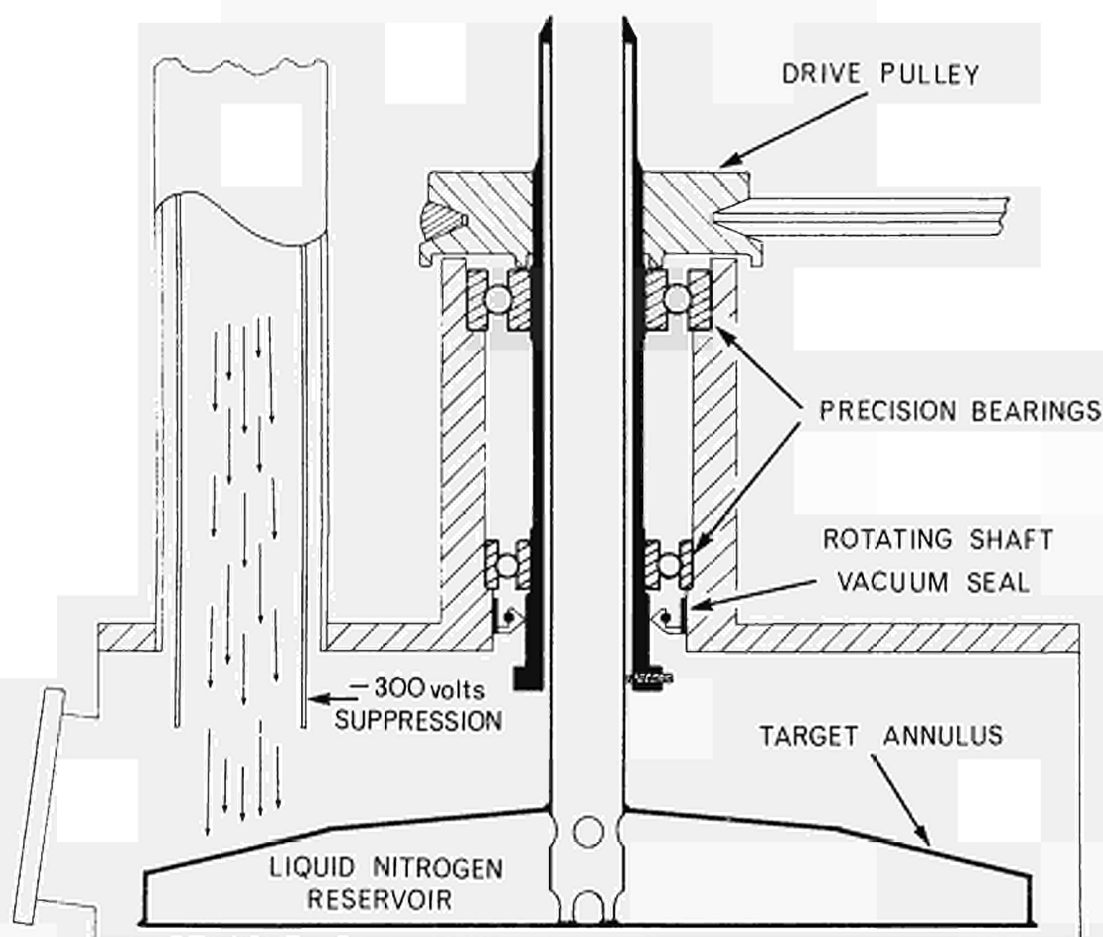


FIGURE 1.

BASIC DESIGN OF ROTATING HEAVY ICE TARGET

LIQUID N₂ ADMISSION.

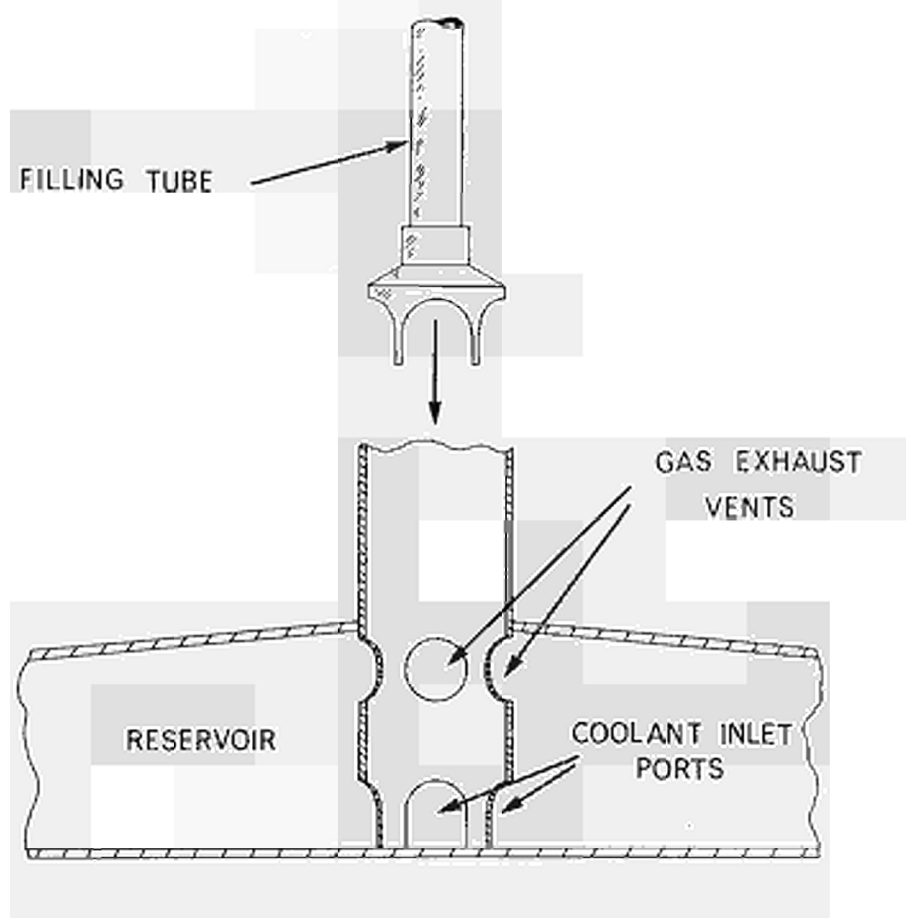


FIGURE 2.

LIQUID NITROGEN FILLING TUBE

D₂O DISPENSER

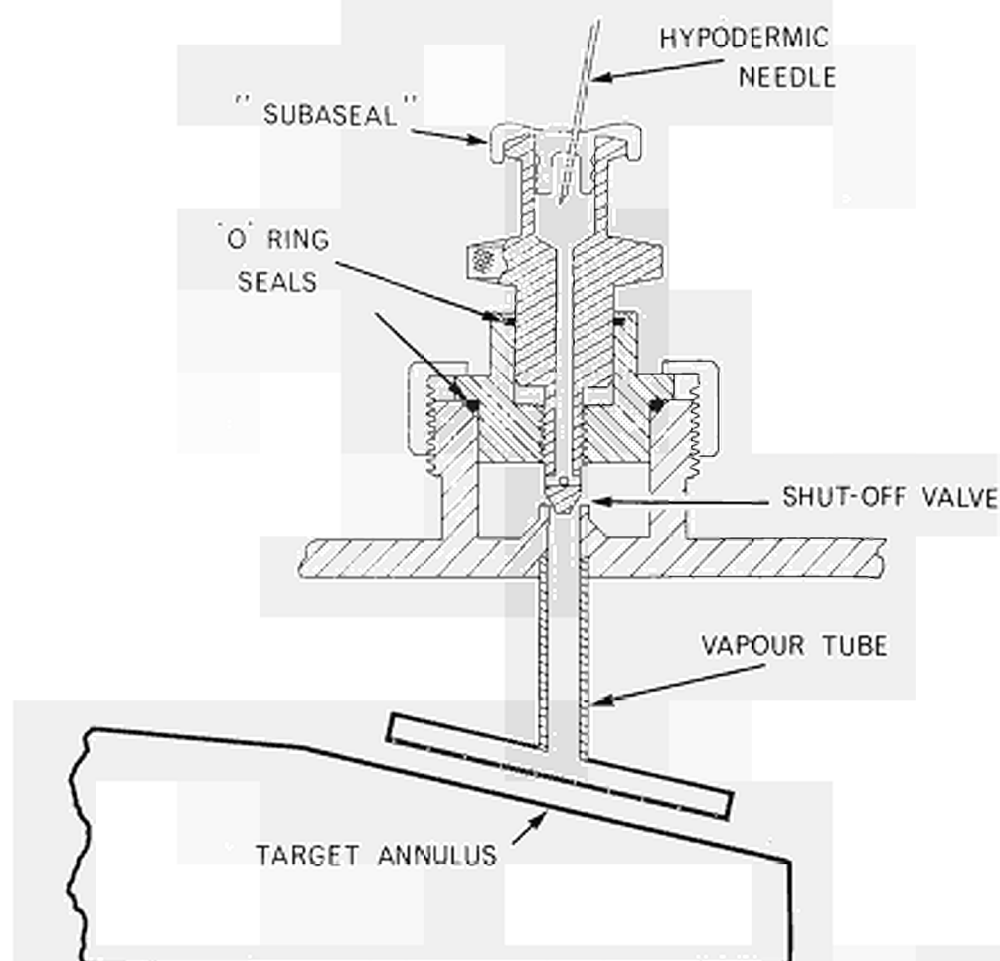


FIGURE 3.

D₂O DISPENSING UNIT

self-sealing rubber plug into the vacuum system. To get an even distribution of the D_2O onto the target surface the vapour is fed through a small bore tube to holes adjacent to the rotating target surface. As a further refinement, the "Subaseal" holder when screwed down tightly, seals the vacuum system. This is a safeguard which allows continuation of use of the target in the event of the "Subaseal" leaking.

4. Performance

Typical conditions which have given stable and reproducible outputs so far have been:- beam currents of the order of 250 μA at accelerating voltages of 600 KV. The beam, which had a core size of about 0.5 cm. was scanned over the target to give an effective target size of 3 cm. x 3 cm. Scanning was accomplished with two pairs of parallel deflector plates, each pair being connected to an oscillator with a frequency of around 100 Kc/sec. Experience has shown that an application of 0.3 cc of heavy water is sufficient to give the maximum output from the target. This quantity gives a target thickness of 12 μ if 100% transfer is assumed, the total target area being 39 sq. in. Volumes in excess of this tend to give higher running pressures without improvement in output. Stable conditions are maintained with pressures of 1×10^{-4} Torr at the target.

The consumption of the He^3 gas for beam currents of the order of 250 μA is about 100 cc per hr.

The results obtained so far have been very encouraging; it has proved possible to obtain stable running conditions with high proton outputs for many hours.

It has been found, however, that a new target requires "conditioning" before high powers are used. This necessitates a steady build-up of beam on the target. Experience has shown that 15 mins to 20 mins is sufficient time to accomplish this. Failure to do this will give excessive pressures in the system, and consequently loss of target.

The reason for this phenomenon is not fully understood but it seems likely that the ice layer when first layed down is of a crystalline structure, with high peaks having low thermal conductivity. In the conditioning stage the ice layer may become homogeneous and uniform

in thickness and the heat transfer probably improved.

The proton yields so far obtained were 1.16×10^{10} protons/sec. for 250 μ A at 600 KV. This compares favourably with an optimum yield obtained from a Ti D. target of 3.48×10^9 protons/sec. at 385 μ A and 600 KV

Acknowledgements

The construction of this rotating ice target was suggested by Dr. G.J. Neary. Thanks are due to Ir van Darsten of Philip Research Laboratories for information about some of the characteristics of such a target, and to Mr. Cossutta of Multivolt Ltd., for valuable discussion of some of the constructional problems.

I am indebted to Dr. G.J. Neary for his advice, to Mr. A. Stretch for his expert technical assistance and to Messrs. R. Wilkinson and D. Martin for their help in the development work.

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"A ROTATING TARGET ASSEMBLY FOR 10^{12} NEUTRONS/SECOND"

D. COSSUTA

Multivolt Limited, Crawley / Sussex, U.K.

SUMMARY:

A new high flux 14 MeV neutron source is described which was specially designed for use in fast neutron radiotherapy and for all applications requiring long irradiation times at a flux of the order of 10^{12} n/second. The design is based on a continuously rotating annular target assembly and is intended for use with accelerating machines providing several milliamperes at energies of some hundreds of kilovolts.

Thermal effects and problems of heat dissipation are discussed in particular reference to the continuously mobile target surface.

The parameters involved give useful information regarding performance which is expected from the present construction. Future development of this design is also briefly discussed.

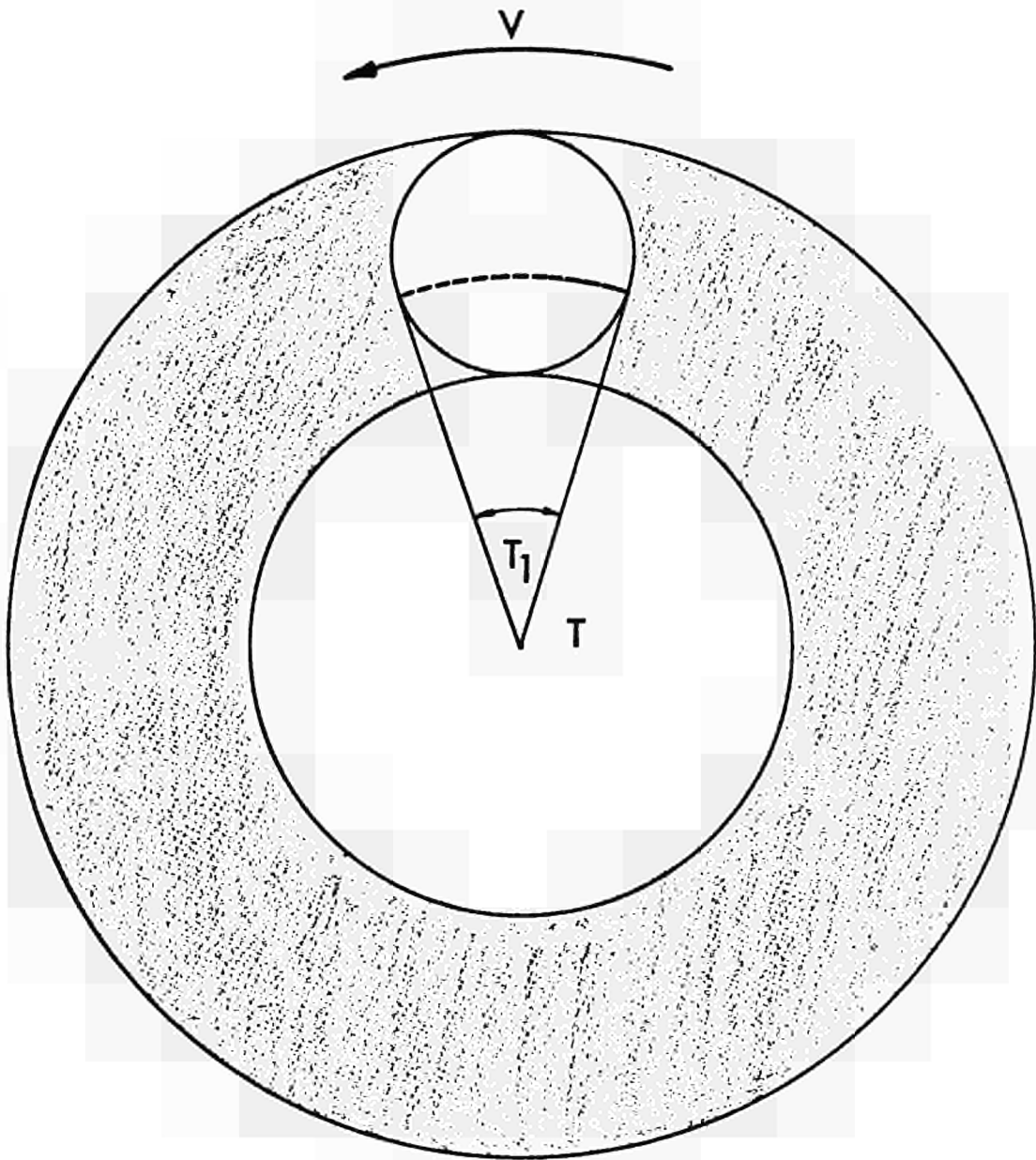
There is considerable interest at present in using 14 MeV neutrons for radiotherapy purposes. I will describe my method of designing a target which it is hoped will provide a useful source for this purpose.

Low energy accelerators capable of supplying a deuteron beam of 2-3 kW power at energies of several hundreds of kilovolts are presently available from various manufacturers. It seems, however, that it is not so simple to dissipate such a power at a stationary tritium target of usual type ^{dimensions} and / without impairing its quality and performance. Much work has been carried out on many aspects on the design of target systems for high yields with the result that a wealth of information is presently available on the nature of tritium targets, on the processes occurring at the target under bombardment, and on the best operating conditions for a long target half-life. At present it is comparatively simple to maintain a neutron flux of the order of 10^{10} n/s during a period of hundred hours, but it seems to be more difficult to maintain a flux of 10^{12} n/s during a similar length of time. A long-life neutron source giving 10^{12} n/s is of particular interest in fast neutron radiotherapy, but also in activation analysis and other applications requiring highest possible neutron flux and long irradiation times. Sealed-off accelerator tubes as described by J. L. D. Wood are presently being developed by some establishments to meet this requirement. For the same purpose a specially designed target assembly for 10^{12} n/s would be a valuable accessory for high power accelerators. To achieve a target half-life of several tens of hours a design based on a large area rotating target is essential. In this paper we shall discuss

heat transfer parameters and thermal effects which occur at the target with particular reference to a continuously rotating target surface. I will also describe a construction which has been developed for the purpose of experimental work, and mention some future development.

In the diagram shown in Fig. 1. the target is represented by the annular surface area which rotates in the direction shown by the arrow. The white circle at the top represents the diameter of the ion beam. If the target rotates at 1000 rpm each surface element along the path shown dotted, will spend 6 msec. under the beam, and its temperature will be increasing during this time. During the remaining part of the cycle the surface will be cooling off. If we consider the dimensions of commercially available annular tritium targets of 146 mm O. D. and 76 mm I. D., the beam diameter is 34 mm and the beam area is 9 cm^2 . Assuming uniform beam distribution at 3 kW total power this gives a specific load of 330 W/cm^2 .

Let us now consider thermal effects occurring at the target during a heat pulse of 6 millisecc. duration, and a specific load of 330 W/cm^2 . When a beam of deuterons strikes the titanium target, the heat will be released in a surface layer equal in thickness to the range of deuterium ions. The time in which this surface layer will reach a given maximum permissible temperature T_{max} , can be calculated for instance by a method as used by H. J. Spaa,¹. It is assumed here that the heat conduction is absent. For a temperature difference of 100°C between T_{max} and the temperature of the cooling medium, values of specific heat and thermal conductivity coefficients for titanium hydride, and aforementioned specific load of 330 kW/cm^2 , we find that T_{max} is reached in about 10^{-5} sec. This parameter is of particular interest in a problem as examined by Spaa, where heat transfer properties of D_2O -ice target were studied in connection with a rotating target system. In the latter case, the cooling-off



ROTATING ANNULAR TARGET

Fig.1

time of the surface layer due to thermal conductivity is two orders of magnitude slower, than the time taken to heat the surface to its maximum permissible temperature. Consequently, a beam scanning system was used in addition to the rotation of the target disc, in order to operate at a safe permissible specific load for D_2O -ice. Identical expressions are given by both, Spaa and Hulek ², for calculating the parameter responsible for heat removal due to thermal conduction. The above mentioned data on the D_2O -ice target are given for comparison with tritiated titanium targets. In this case, the time for the temperature of the surface layer to decrease by an order of magnitude is about 10^{-7} sec. Clearly, in the case of titanium tritide, the temperature of the surface layer decreases by two orders of magnitude faster, due to the heat transferred to the lower layers and into the base. In practice, however, the surface layer is subjected to many adverse effects, such as sputtering, deposition of a carbon layer, non-uniform beam distribution etc., which may well change the picture of the heat removal from the surface.

If we now assume that the temperature gradient between titanium tritide and copper base is not appreciable, which seems to be true for good quality targets, we can observe further the process of heat transfer into the volume of the copper base. In fact, a heat pulse of 6 millisec. duration and specific load of $330W/cm^2$ is sufficiently long to produce an increase in the temperature of the base, and establish a certain temperature gradient through the volume of the base metal. Since the titanium hydride is very thin one can assume that the target is not composite, but uniform, with thermal properties of copper throughout the thickness. It is then possible to calculate the maximum permissible power input during which the surface temperature will not exceed T_{max} . For this purpose one can use Carslaw's expression, or a similar formula given by Hulek "for an infinitely thick plate whose surface is suddenly subjected to a thermal load".

Carslow's expression gives a value of 730 W/cm^2 for maximum power input for a heat pulse of 6 millisecc. during which the surface temperature rises for 100°C . If the target disc were rotating at a rate of 10 rpm, this would correspond to a heat pulse of 0.6 sec. and the maximum permissible specific thermal load would be 73 W/cm^2 . In the discussion following this paper Mr. Laverlochere of Centre d'Etudes Nucleaires de Grenoble will give some data obtained from a recent experiment with a 10 rpm rotary target. These results are in agreement with the calculated values, and they indicate that a total dissipation of 3 kW can be achieved at 1000 rpm. In fact, the initially mentioned requirement to transfer a thermal load of 330 W/cm^2 from an area of 9 cm^2 to the cooling water, in order to obtain 10^{12} neutrons/sec. is not too difficult to achieve. Specific loads of this order can be dissipated in stationary target holders of usual design, while considerably higher dissipation is possible by more complex designs as for instance described by Rethmeier and Van der Meulen³. They have established that the most critical is the heat transfer from the target base to the cooling medium, and that the best method is to use a system of cooling fins with an adequate water velocity.

Figure 2 shows the design of a rotor of a Multivolt rotating target. Titanium tritide is formed directly on the copper disc, so that soldering is avoided. Two large diameter concentric tubes, which also form the shaft of the rotor, are intended for a water throughput of 20 lit/min. On the inside of the copper disc a number of grooves are machined radially to form cooling fins, essential for an efficient cooling. The plate behind the actual target area is 4 mm thick in order to serve as an efficient heat sink. This brings us to a feature of continuously rotating targets which was studied by DuMond et al⁴ in connection

with continuously mobile X-ray focal spots. If we refer again to Fig. 1 we see that during continuous rotation, a "steady" state of thermal oscillations is set up. At infinite speed of rotation the surface temperature will be the same for all surface elements at the same diameter, and the total load will be more or less uniformly spread over the entire ring. Consequently, for the case under discussion, where $T/t_1=10$, ten times as much power could be safely applied to the disc, as compared with a safe load for a stationary spot. In this study the flow of heat was assumed to be one dimensional and perpendicular to the target surface. This would correspond to a hollow target carrying plate. From the data given by DuMond, at 1000 rpm about 4 times as much power could be applied to a 5 mm thick rotating target disc. For the target disc in Fig. 2, the heat is spreading laterally, and there is a gain due to thermal storage in the copper.

Fig. 3 shows the cross-section of the small rotating target which was developed in order to test the design considerations described in this paper. In this unit the vacuum rotary seals and driving mechanism is substantially different to the one employed in the 10 rpm Model. In fact, the present construction is substantially different from the diagram in Fig. 3, as double rotary seals and two ball bearing races were used in the development model. The beam entry tube is at 45° with respect to the target plate which decreases the specific load due to the beam for a factor of about 1.6. One of the major problems with an assembly for neutron radiotherapy is that the target will need replacing regularly by some person in hospital, and due to the high level of induced activity, the target changing should be swift and simple. As regard to the half-life of this target, this can be calculated from the experimentally obtained values reported in the literature. By taking a value of $3 \text{ mA}\cdot\text{hr}/\text{cm}^2$, which it seems can be achieved with good quality tritium targets, and good vacuum conditions,

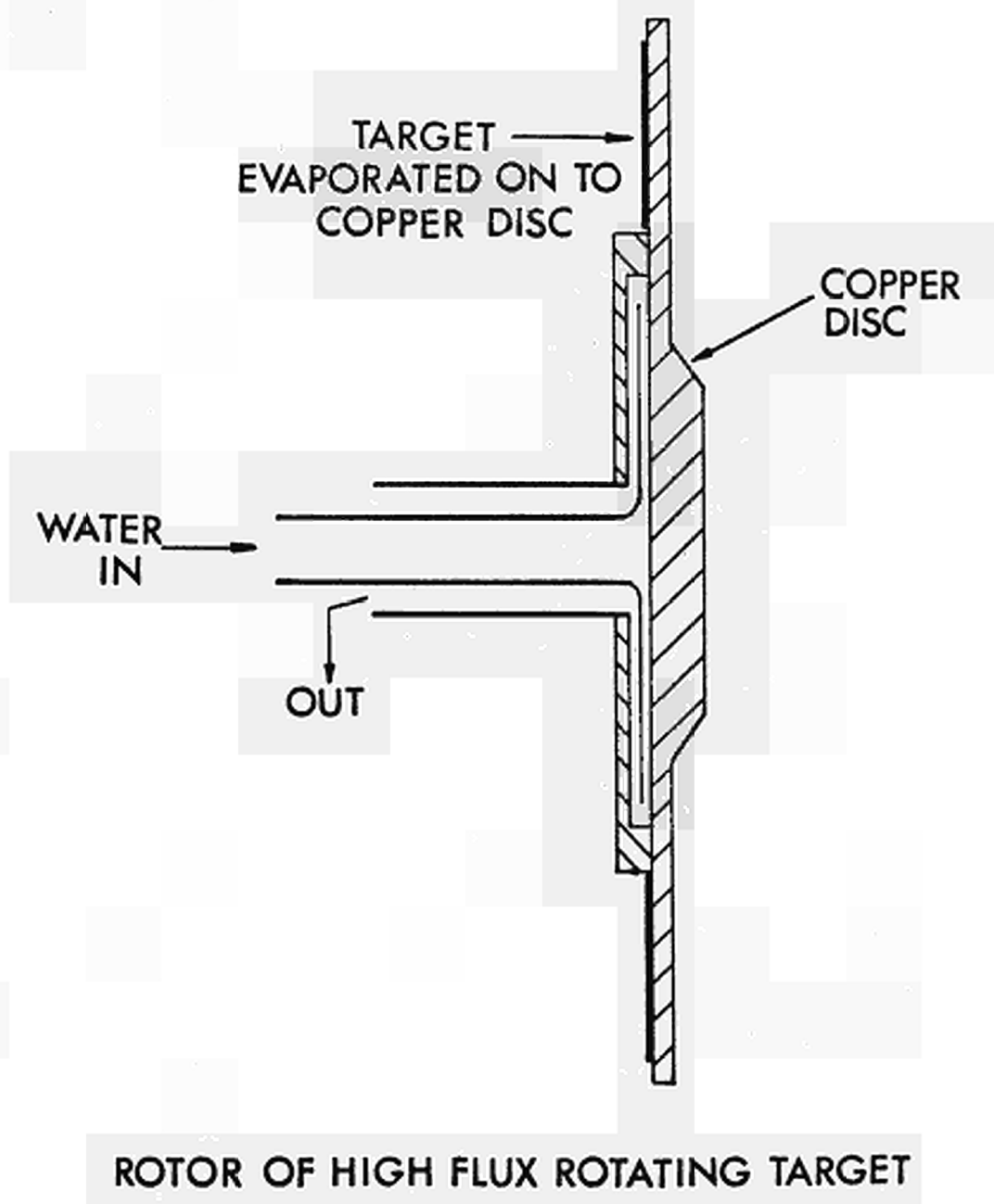


Fig.2

Rotating Target Holder . type RTH2R for High Flux Fast Neutron Radiotherapy - Model 9

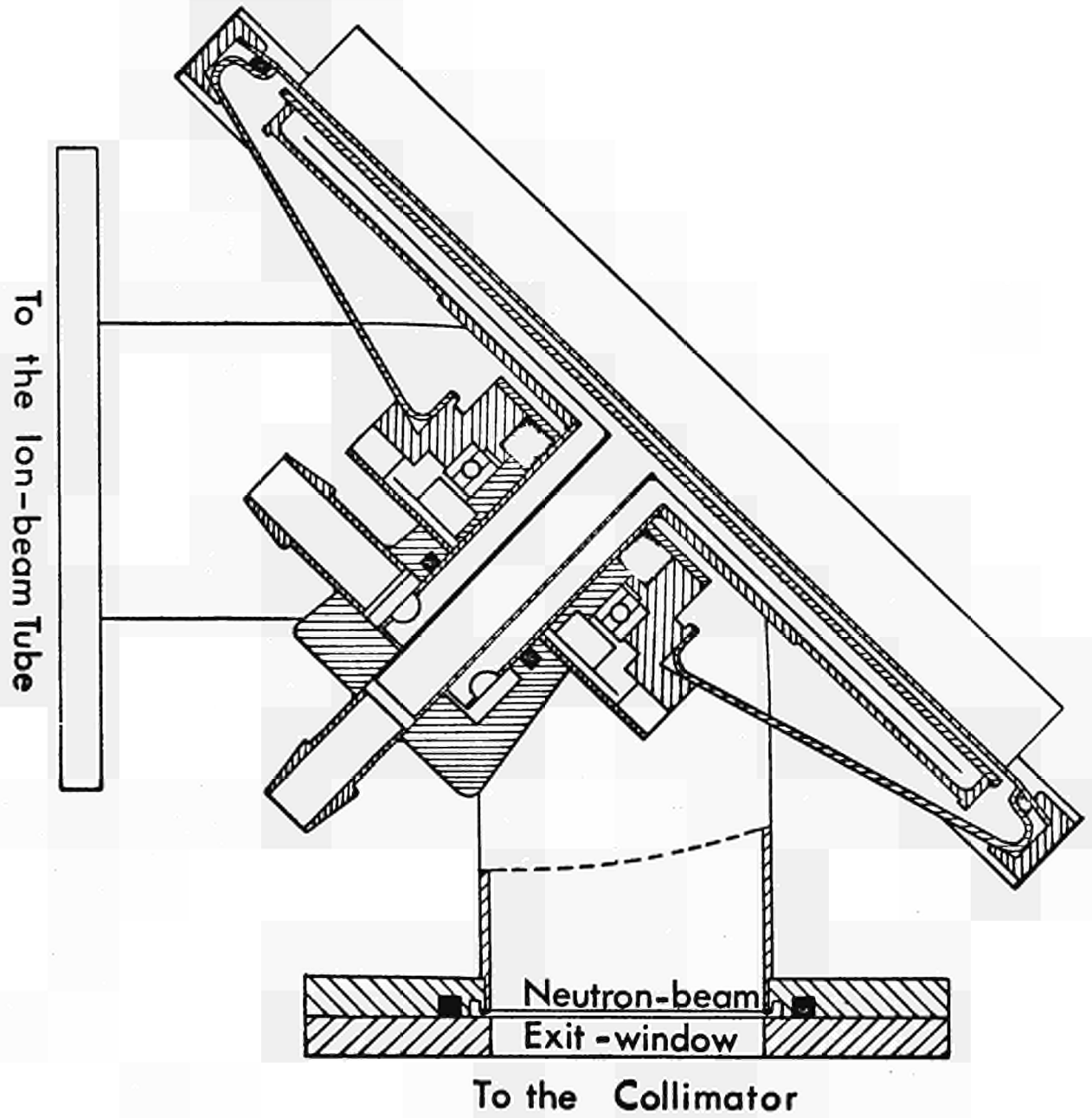


Fig. 3

a half-life of 36 hours is expected from an annular target of 121 cm^2 . If a longer half-life is required a larger area rotary target could be constructed. To accomodate a larger area target within the present design it would merely be necessary to increase the diameter of the vacuum enclosure, and that of the target disc.

Major problem would then be for the target manufacturers to fabricate greater area annular targets, or sections of an annular target of greater diameter. To conclude this paper let us recall some data of a rotating target based on the use of sections to form a large area target, as reported by Mr. Smith⁵ of Aldermaston. This assembly is shown in Fig. 4 of his paper. The target itself is 20 cm radius, 5 cm wide, total area 650 cm^2 containing 2000 curies of tritium. At 300 kV and 10 mA the neutron yield was 10^{12} n/s . If we used a target with a half-life of 3 mA.hr/cm^2 , this amounts to a half-life of 1800 hours.

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DISCUSSION

Mr. WOOD

Mr. COSSUTA quoted a predicted half life of 1800 hours at 10^{12} n/sec for a 650 cm^2 target, based on a figure of 3 mA hr/cm^2 . Surely this is a half life in mA hours and must be reduced in proportion by the value of current needed to obtain the output of 10^{12} , e.g. at 250 keV at least 10 mA and a life of only 180 hours. What are his comments ?

Mr. COSSUTA

The total surface area is 650 cm^2 , and if one assumes that the figure of 3 mA h/cm^2 is correct that would give a half life of 1800 h.

Mr. RICCI

Have you obtained the figure of 10^{12} n/sec by actual measurement ?

Mr. COSSUTA

The rotating target assembly for neutron radiotherapy described in this paper has not been tested with an accelerating machine as yet. Up to the present date it was only tested in operation under vacuum. It is hoped, however, that the testing of the target life at 10^{12} neutron/second will be carried out in the near future.

Mr. LAVERLOCHERE

Nous avons essayé le dispositif MULTIVOLT type RTH2B, dans le but de mesurer la $\frac{1}{2}$ vie de deux cibles différentes. Conditions opératoires : 350 kV 500 μA , \emptyset faisceau 25 à 27 mm, mesure de flux par disque de cuivre de diamètre 10 mm. Les résultats ont été les suivants :

- 1) cible à refroidissement central (25 curies ^3H) : cible vissée.
Flux au début de l'essai $2,6 \cdot 10^8$ n/cm²/s, ce qui correspond à une émission de $9,1 \cdot 10^9$ n/s dans 4π 1/2 vie de l'ordre de 60 heures (essai continu de 38 heures). La puissance maximum admissible par ce dispositif a été mesurée égale à 350 kV - 700 μ a.
- 2) cible à refroidissement arrière (82 curies ^3H) : cible soudée.
Flux au début de l'essai $1,9 \cdot 10^8$ n/cm²/s, ce qui correspond à une émission de $6,7 \cdot 10^9$ n/s dans 4π 1/2 vie de l'ordre de 150 à 200 heures (essai continu de 30 heures). Le rendement neutronique obtenu est donc de l'ordre de $2 \cdot 10^7$ n/s/ 4π par microampère.

Mr. MORGANSTERN

I wish to bring to the attention, recent work on high power targets at Cornell University; these are stationary, water cooled targets handling up to 4 to 5 kW/cm². This work is being pre-formed by Peter HANLEY at Cornell improving upon the original work by Harry WEGNER at Brookhaven National Laboratory.

Mr. WOOD

Could I ask Mr. MORGANSTERN for further details of the target that has been operated at 4 to 5 kW/cm² at Cornell ?

Mr. MORGANSTERN

Work was described by WEYVER at the Accelerator Conference in Washington on 1965. Depends upon a squared copper tube with capillary providing turbulent flow between target material base and wall of copper tube. The theoretical calculation indicated 7 kW/cm².

Mr. WOOD

Thank you. This encourages me in the development of the sealed tube for 10^{12} neutrons/second which I was describing yesterday.

Mr. FABIAN

Ich möchte Herrn LAVERLOCHERE fragen, warum das rotierende Target von Herrn COSSUTA bei der Belastung von 1 mA und 250 kV nur die Ausbeute von $9,1 \cdot 10^9$ n/sec ergab. Das finde ich viel zu wenig. Um den Faktor 5

sollte die Ausbeute höher liegen. Welches Atomverhältnis H/Ti hatte diese rotierende Scheibe ?

Mr. LAVERLOCHERE

Je n'ai pas d'explication au sujet du faible rendement obtenu effectivement au cours de ces essais. Bien sur, différentes hypotheses peuvent être suggérées, mais il serait avant tout important de savoir si d'autres laboratoires ont essayé ce dispositif et quels résultats ils ont obtenus. Nous avons suivi strictement le mode opératoire prescrit. Peut-être existe-t-il des précautions spéciales à prendre, mais nous ne le savons pas.

Mr. BROERSE

Starting from the emission data given by Dr. LAVERLOCHERE, i.e. $9 \cdot 10^9$ n/sec for a 350 kV high voltage and a 500 μ A deuteron beam current, it has to be concluded that for an output of 10^{12} n/sec extremely high currents have to be employed. Normally it is expected that the output of 10^{12} n/sec can be achieved at an ion current of 8 mA. For the COSSUTA target, however, this current should be equal to 50 mA. The low yield of your rotating targets does not give any reason for optimism in constructing a 10^{12} n/sec source with an acceptable target half life.

Mr. COSSUTA

The practical values of ion-beam parameters for a neutron flux of 10^{12} n/s seem to be 10 mA at 300 kV. Consequently, I would hesitate to calculate any such parameters from the data given by M. LAVERLOCHERE as it seems that at present their measurements are not considered as final. On the other hand, neutron flux figures given by Mr. SMITH and other users of Multivolt Rotating Assembly indicate that the target performance in a rotating assembly is of the same order as in stationary holders of usual design.

Mr. SMITH

Comment on the discussion of target life variation.

I have achieved a neutron source strength with my machine of $2 \cdot 10^{12}$ n/s for a period of a few minutes. Since I had no required for this output I did not try to maintain it. However at lower currents I have maintained a neutron

source strength of $8 \cdot 10^{11}$ n/s over a period of 6 hours with ion currents of 5 to 8mA. I used a 5cm diameter static target for this purpose.

I believe the difference between neutron yields obtained with rotating and static targets is probably due to bad handling of the target. I have had difficulty in soldering target rings on to rotors. I believe that if manufacturers make targets directly on to the rotors then these difficulties can be overcome and adequate neutron yields will be obtained with ion currents of around 20 mA with either type of target.

Mr. FABIAN

Die Herstellung von rotierenden Targetscheiben ist wesentlich schwieriger als die Herstellung von kleinen Cu-Targets. Das Massenverhältnis von Cu zu Titan und die Drehbearbeitung des Cu-Flansches bringt unvermeidlich Unreinheit in das Hochvakuum beim Erhitzen der Cu-Massen. Dies hat zur Folge, dass das Atomverhältnis von 1,5 H/Ti, wie es bei kleinen Targets vorliegt, nur mit wesentlich grösserem Aufwand erreicht werden kann.

Aber das Target von Herrn COSSUTA war kein bedampfter Flansch sondern ein aufgelöteter Cu-Bleching. Es wäre denkbar, dass beim Auflöten des Ringes auf den rotierenden Flansch ein Teil des Tritiums verloren ging. Dies könnte eine Erklärung der geringen Neutronenausbeute beim Versuch sein.

Mme. BREYNAT

La pression à l'intérieur de la cible a certainement beaucoup d'importance. La section du pompage sur la boîte à cible est faible.

Si la pression partielle d'oxygène augmente, à 180°C environ, température atteinte par la cible sous l'impact du faisceau, il y a sûrement oxydation du titane. La présence de joints torriques en perbunan assurant l'étanchéité du porte cible le long de l'axe tournant, entraîne sûrement la pollution de la cible. Ces raisons contribuent à diminuer le rendement neutrogène de la cible. Lorsque nous voulons utiliser nos cibles dans de bonnes conditions, nous sommes obligés d'utiliser des techniques " d'ultra vide " pour assurer l'étanchéité de la fixation la cible sur l'axe.

CIBLES NEUTRONIGENES TRITIÉES OU DEUTÉRIÉES

M. DETAINT

C.E.A. - C.E.N./G. SECTION DES ACCELERATEURS
GRENOBLE - FRANCE

Je vais vous présenter, brièvement, quelques résultats d'expériences que nous menons actuellement à Grenoble, dans le cadre de recherches consacrées à l'amélioration des cibles neutronigènes tritiées ou deutérées.

Le but de ces essais est d'étudier le comportement sous faisceau des dépôts en couches minces des différents métaux utilisés pour la fabrication des cibles, ceci afin de déterminer, dans chaque cas, l'importance des différents facteurs limitant la période des cibles.

Nous cherchons donc, dans ces expériences, à tester la résistance des dépôts (adhérence), à évaluer la pulvérisation cathodique (sputtering) due aux deutons et à mesurer l'enrichissement en deutérium.

Les résultats que nous avons déjà obtenus recoupent d'ailleurs ceux qui vous ont été présentés par plusieurs orateurs.

Dans ce court exposé, je vous parlerai surtout de l'enrichissement en deutérium d'un film métallique, c'est-à-dire du problème des autocibles; en effet, il y a là une possibilité d'obtenir simplement des quantités appréciables de neutrons par réaction $d(d,n)$.

Nos expériences sont faites avec un courant de faisceau de 1 mA à 300 KV, le diamètre du faisceau étant de 25 mm. Le porte-cible, à refroidissement annulaire, permet d'avoir une température maximale de 100° C. au centre de la cible.

Nous avons d'abord considéré des autocibles utilisant des métaux qui ne donnent pas d'hydrides stables (au sens thermodynamique du terme).

La FIGURE 1 montre, en fonction de la charge reçue, le débit produit par des autocibles en or, argent et chrome.

On remarquera que le débit 4π est de l'ordre de $5 \cdot 10^8$ neutrons/sec.; au départ, les courbes d'enrichissement sont irrégulières, vraisemblablement à cause d'impuretés superficielles.

La perte de métal par sputtering est sensiblement nulle pour le chrome ; elle est forte pour l'argent et l'or :

$$\begin{array}{ll} 8 \cdot 10^{-2} \text{ mg/cm}^2/\text{coulomb} & \text{pour l'or} \\ 2,7 \cdot 10^{-2} \text{ mg/cm}^2/\text{coulomb} & \text{pour l'argent} \end{array}$$

Pour le cuivre, nous trouvons des valeurs du même ordre de grandeur que pour ces derniers métaux.

Les métaux, dont les hydrures sont très stables, permettent de réaliser des autocibles qui produisent beaucoup plus de neutrons que les précédents.

La FIGURE 2 donne les courbes d'enrichissement de couches minces de titane ou d'yttrium. Les débits obtenus sont de l'ordre de quelques 10^{+9} n/s. 4π . On remarquera qu'à épaisseur égale, le titane est moins enrichi que l'yttrium. La décroissance des débits de neutrons produits résulte de la diminution de l'épaisseur de la partie active de la cible.

La FIGURE 3 donne les courbes d'enrichissement de cibles à base d'ytterbium, d'épaisseurs voisines de 1 mg/cm^2 . Les débits de neutrons 4π sont de l'ordre de $5 \cdot 10^9$ n/s. Ces cibles sont réalisées par dépôt successif d'un métal peu hydrurable sur un support d'argent, puis de l'ytterbium. Pour les cibles considérées sur cette figure, les sous-couches utilisaient de l'or, du chrome ou du cuivre.

L'adhérence de l'ytterbium est différente sur chacun de ces métaux.

La Photographie N° 4 montre l'aspect de la cible d'ytterbium sur or, support argent ; après 6 h 30 de faisceau, elle est détruite bien que son débit de neutrons soit encore important: $5 \cdot 10^9$ n/s dans 4π

La PHOTOGRAPHIE N° 5 montre l'aspect de la cible d'ytterbium sur chrome, support argent, après bombardement. Elle est restée adhérente, mais son épaisseur a diminué par suite de la pulvérisation cathodique.

Je voudrais, avant de conclure, vous dire un mot concernant le choix de l'ytterbium comme matériaux de cible.

Comme vous le savez, l'ytterbium est un métal de terres rares qui se distingue quelque peu des autres membres de cette famille. Son hydrure est très différent de ceux des métaux voisins (en particulier en ce qui concerne la nature des liaisons hydrogène-métal).

Il possède un certain nombre de propriétés dont les plus intéressantes sont : sa très grande stabilité thermique et le fait rare que l'hydrure d'ytterbium est moins volumineux que le métal.

Comme conclusions provisoires de cette étude, que nous poursuivons,
nous dirons que :

- 1.- les autocibles offrent la possibilité de produire simplement des flux de neutrons appréciables,
- 2.- l'ytterbium est le meilleur des matériaux d'autocible que nous ayons essayé (parmi Y Ti Er Zr)
- 3.- les débits obtenus avec des autocibles constituées par des métaux ne donnant pas d'hydrures stables sont également importants. Si l'on utilise des cibles trop minces de titane tritié, on risque une importante production parasite de neutrons de 3 MeV.

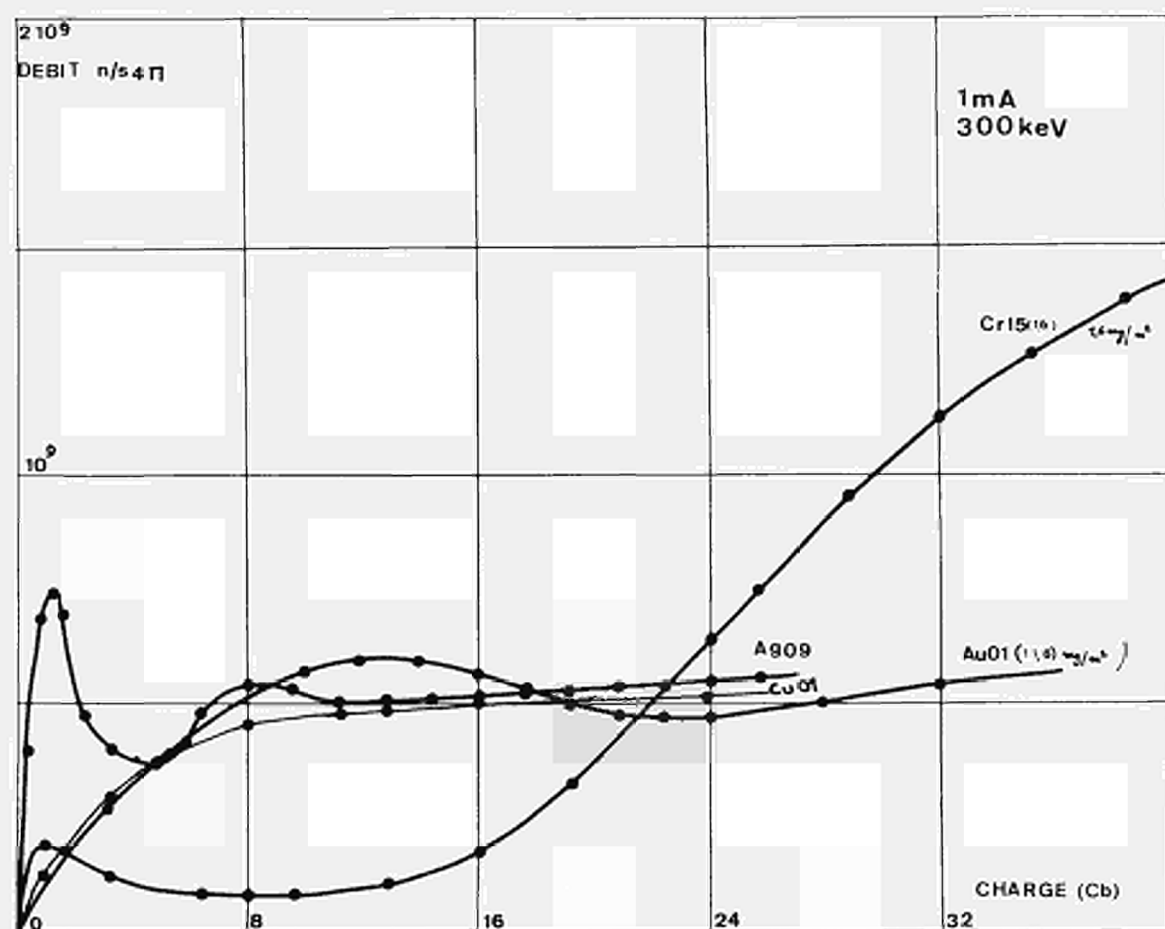


FIGURE 1

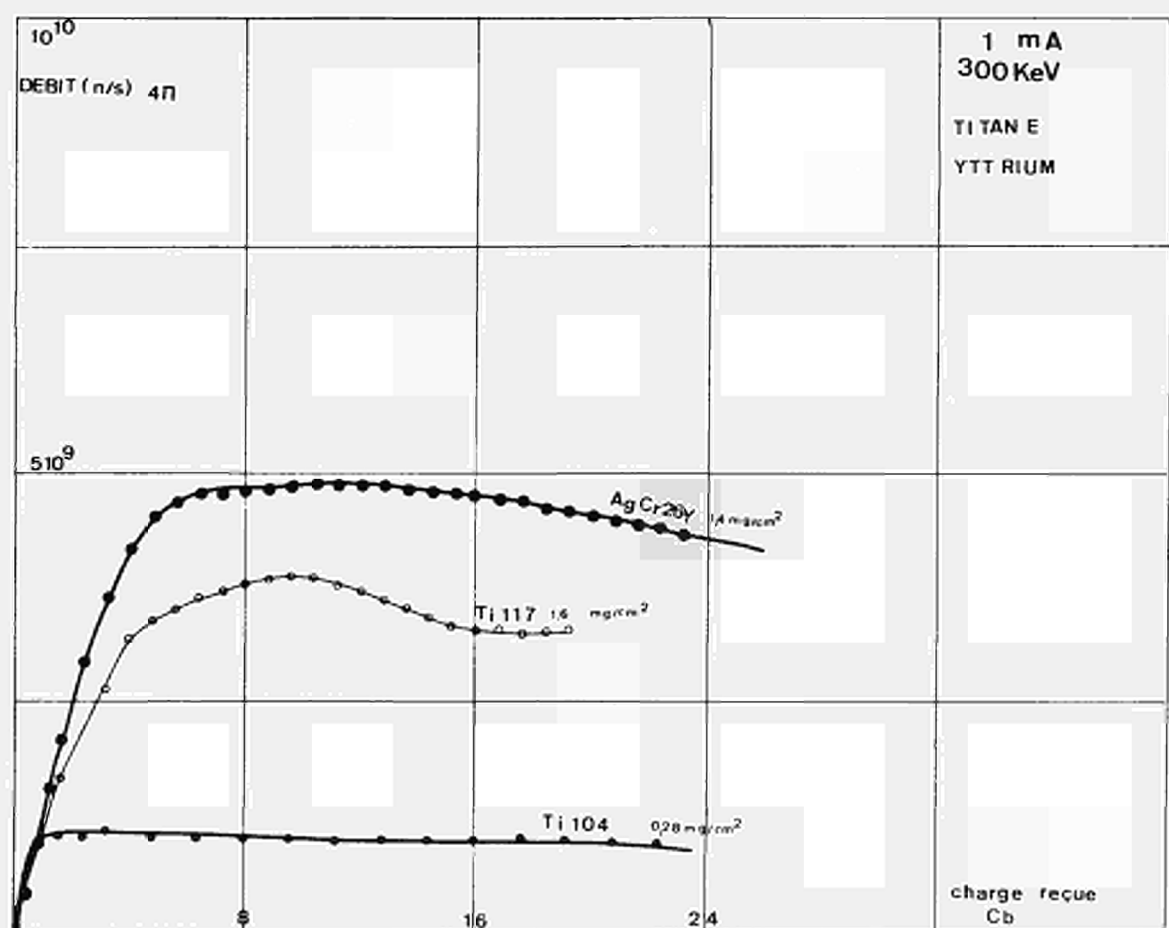


FIGURE 2

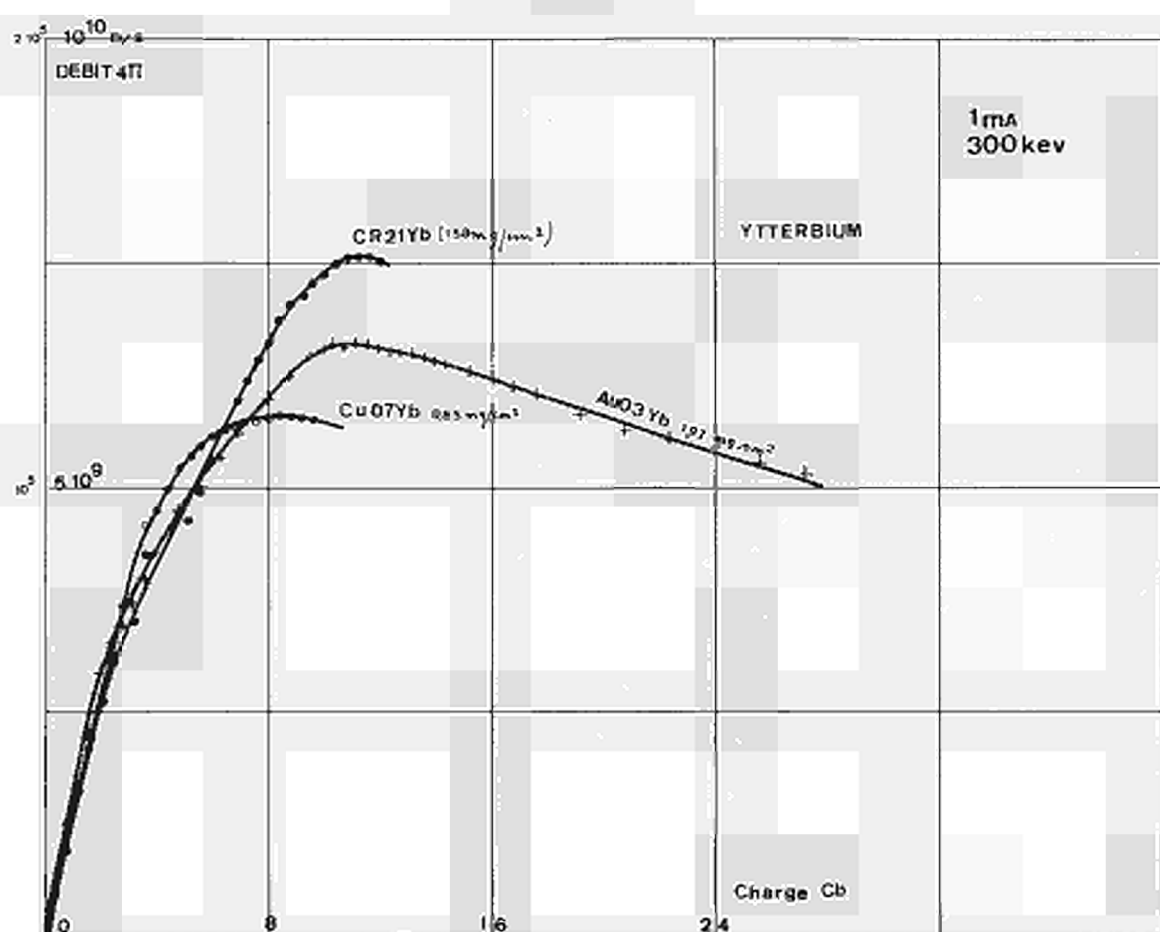


FIGURE 3

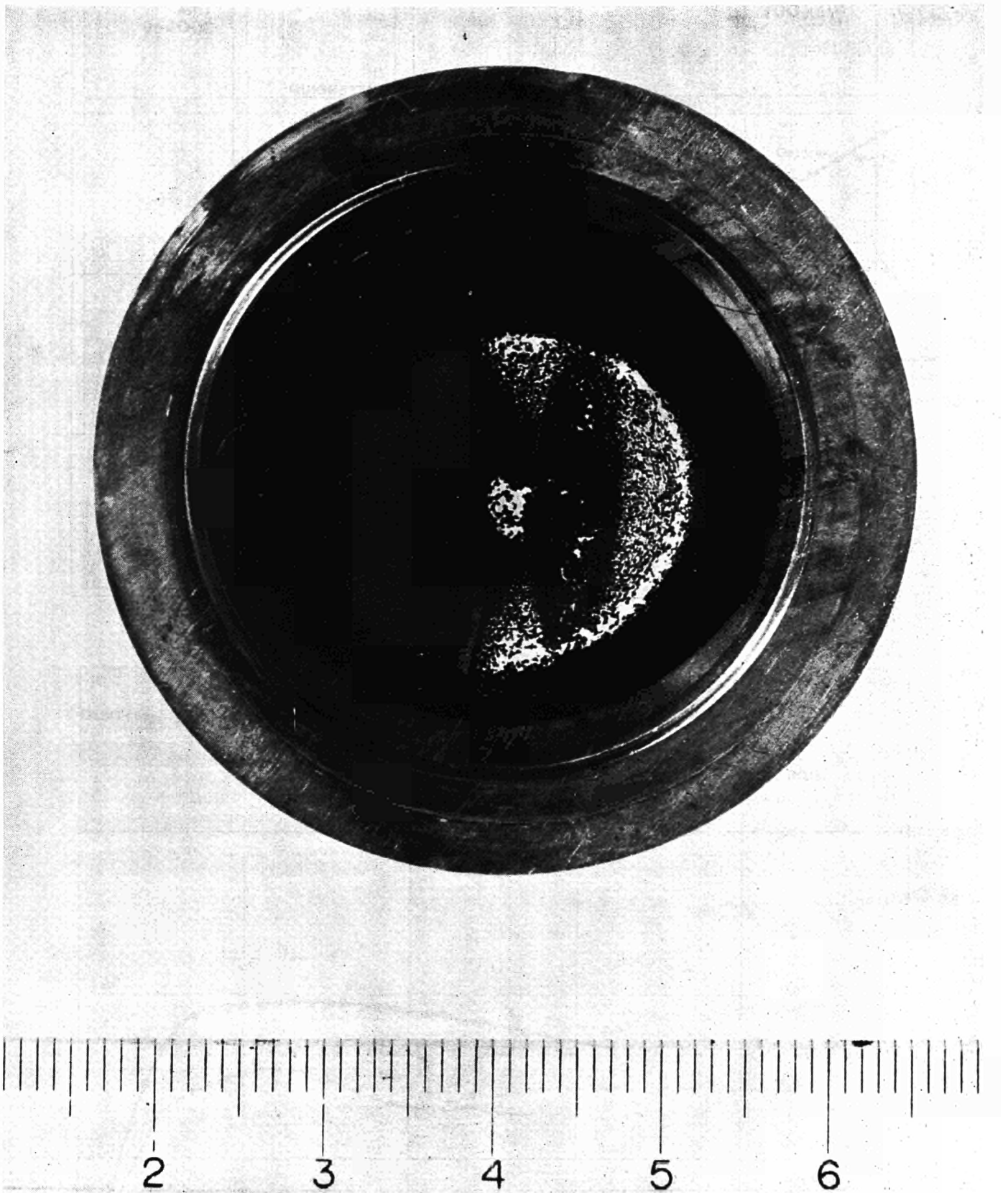


FIGURE 4



FIGURE 5

DISCUSSION

Mr. FABIAN

Ich habe 3 Fragen, wie hoch ist die Neutronenausbeute verglichen mit Titan, wie gross ist die Lebensdauer verglichen mit Titan und wie gross sind die Lebensdauern für andere seltene Erden?

Mr. DETAINT

Le rendement théorique, à épaisseur égale et à richesse égale des cibles de titane deuterées ou tritiées est supérieur à celui des cibles utilisant les métaux de la famille des terres rares proprement dites. Cependant rien n'empêche, sinon la nécessité d'une fabrication particulièrement soignée, d'employer des cibles plus épaisses de métaux des terres rares car l'énergie disponible avec la majorité des machines permet une pénétration des deutons très supérieure à celle que l'on utilise réellement avec les cibles titanes minces de fabrication courante. On ne doit pas également oublier que la stabilité thermique des hydrures de terres rares à composition égale est très supérieure.

Vous dites que les cibles de scandium (métal voisin du titane au point de vue du numéro atomique) donne des rendements inférieurs au titane malgré la plus grande richesse du tritium de scandium. Ce fait n'est pas très étonnant car la stabilité thermique de l'hydrure de scandium n'est pas aussi stable que celle des autres métaux des terres rares.

Nos résultats concernant des autocibles, ils ont été trouvés reproductibles. Ils semblent bien n'être pas en accord avec les résultats concernant les cibles tritiées. Nous sommes à la recherche d'une explication. Les teneurs en deutérium des films après bombardement atteindraient des valeurs très importantes si tous les deutons sont retenus dans le film métallique.

Mr. FABIAN

Sie beurteilen die Qualität der seltenen Erden, glaube ich, zu optimistisch. Aus unseren Untersuchungen schliessen wir, dass von speziellen Forderungen, wie z.B. Temperaturbeständigkeit, abgesehen das Titan zu bevorzugen ist gegenüber allen anderen Elementen, eingeschlossen auch Scandium.

Background problems in thin heavy ice targets

F. Manero

Junta de Energía Nuclear, Madrid, Spain

ABSTRACT

Thin heavy ice targets have been used extensively as sources of monoenergetic neutrons, specially for total neutron cross section measurements, where it is of the greatest importance the energy resolution of the neutron beam. This energy resolution can be worsened particularly by the background of spurious neutrons, as those produced at the target backing material, and mainly by the neutrons produced at the self-target formed by the loading of the target support with the deuteron beam. In this work the importance of these backgrounds is studied. It is found that the contribution of the neutrons produced by the reaction of the deuterons with the target-support material can be considered negligible and can be omitted in any correction. On the contrary the contribution of the neutrons produced at the self-formed deuterium target is very important, and their number approaches that of the true target neutrons. The growth of the neutron background with bombarding time is studied, and it is shown that it tends to a saturation value which can be reached quite quickly.

Introduction

Thin heavy ice targets have been used for long and extensively by many experimenters as a source monoenergetic neutrons (1-8). This type of target and the reaction $D(d,n)$ constitute a very suitable source of neutrons in the neutron energy range from 3 MeV to 6 MeV, because it can be formed easily, its thickness can be maintained constant through the whole energy range and its neutron production is high enough to carry out neutron cross section measurements in short periods of time. In fact, for the bombarding currents that the target can support without evaporating, the neutron yield is similar to that obtained with other types of deuterium targets.

In any neutron cross section measurement the energy spread of the neutron beam plays a very important role. The background of spurious neutrons produced in any part of the experimental set-up but the target, and in the particular case of the heavy ice target those produced at the target backing, can contribute in an appreciable amount to this energy spread.

In this paper a study of the backgrounds found in the use of

heavy ice target is presented, with a special emphasis on the background of neutrons produced by means of the $D(d,n)$ reaction at the self-target formed by the loading of deuterium on the target-support. This self-target, as already reported by Fiebigler (9), can be very important in some cases.

2. Experimental Methods

The experimental arrangement was that used for the total cross section measurements carried out in this laboratory (8).

The target backing was a thin copper sheet, put at 45° to the beam path, and cooled by liquid air through a hollow copper tube 2 cm in diameter. The heavy ice target was obtained by condensing D_2O vapour on the copper surface. The target thickness was determined by the number of counts on the detectors, and its formation was controlled by means of a needle valve and the evaporation time.

A large surface liquid air trap was placed near the target and cooled every day about one hour before the beginning of the measurements, in order to prevent the formation of carbon deposits on it.

In spite of this precaution it was not possible to avoid, that after long periods of working, a very thin layer of carbon was formed by condensation of residual vapours onto the target from the vacuum system, due to the vapour pump effect of the cold surface of the target.

The self-target background measurements were made in the course of the total cross section determinations, where the deuteron energy was varied from 0.4 MeV to 2.0 MeV by steps of 50 keV and the bombarding time for each energy ranged from 60 minutes at 0.4 MeV to 20 min. at 2.0 MeV. The deuterium beam was kept equal to $5\mu A$ in all the measurements. The size of the beam spot on the target was determined by the focussing conditions and the limiting diaphragms, and was kept of the order of 1 cm^2 .

The neutrons were detected with an Emmerich detector (44 mm diameter and 15 mm thick) coupled to a Dario 53AVP photomultiplier, in order to discriminate against the gamma rays from the reaction $^{16}\text{O}(\text{d}, \text{p} \gamma) ^{17}\text{O}$ in the heavy ice target. The discrimination level was set such that all the pulses due to the gamma rays were rejected.

3. Results

Two kinds of backgrounds can be considered: the gamma-ray background, that determines the choice of the neutron detectors and the neutron background, the really important one from the point of view of energy resolution.

3.1. Gamma-ray background. The main source of gamma rays is the $^{16}\text{O}(\text{d}, \text{p} \gamma) ^{17}\text{O}$ reaction in the heavy ice target, being its Q of 1,919 MeV. The O^{17} has levels at 0,871 MeV, 3,048 MeV and 3,846, that can be reached with deuterons of 2 MeV (10). The 0,871 MeV level is the more important because it is populated in almost the same amount that the ground state and its decay gamma ray gives, on the organic neutron detectors commonly used, a pulse of the same size that those of a 3-4 MeV neutron (11). In any case this gamma-ray background is not a trouble because can be easily removed by pulse shape discrimination technics or a convenient choice of the detector.

3.2. Neutron background. It is possible to distinguish two main sources: The neutrons produced by the deuterons with the structural materials that form or surround the target, and the neutrons produced at the self-formed targets on these materials. We restrict our study in both cases to the target backing because it gives the main contribution. Another source of background neutrons are the limiting diaphragms, but of less importance because they are farther away from the detectors and the self-formed targets on them should be smaller.

The target support was a copper sheet, thick to the deuteron range. The deuteron beam can produce neutrons on the copper through the reactions $^{63}\text{Cu}(d,n)^{64}\text{Zn}$ and $^{65}\text{Cu}(d,n)^{66}\text{Zn}$, with Q 's of 5,4817 MeV and 5,679 MeV respectively (12). The maximum energy of the emitted neutrons, for 2 MeV deuterons, is 7,47 MeV and 7,63 MeV according with kinematics; but their number is extremely low, as shows the fig. 1, where this number has been plotted vs energy. If this number is compared with the yield of a 100 keV-thick heavy ice target, measured in the same experimental conditions and with the same discrimination levels, it is seen that this background is of the order of 0,7%, and does not introduce any appreciable inaccuracy.

These results are in agreement with the corresponding reaction cross sections: ~ 30 mb/sr at 0° for 2 MeV deuterons for the $D(dn)$ reaction (13) and $\sim 10^{-2} - 10^{-1}$ mb/sr for the $\text{Cu}(d,n)$ reaction (14).

Much more important than the neutrons emitted by the backing material are those produced at the self-target formed by the loading of deuterium from the deuteron beam. These neutrons are of the same kind that the target neutrons but they can show a larger spread in energy depending of the depth reached by the deuterium in the backing.

To know the importance of this self-formed target, we have studied its dependence with the charge deposited on the target backing. The results obtained are plotted in fig. 2 and 3.

The fig. 2 shows the dependence of the neutron background with energy after several bombarding times. The deuteron beam was ever equal to $5\mu\text{A}$, the time is given in minutes. The working conditions of the detectors are the same as in fig. 1. The fig. 3 shows the number of counts at the detector as a function of the bombarding time and the deposited charge. Both sets of curves show that the contribution of the self-target neutrons can be very important; it tends to a saturation value, that can reach almost a 30% of the yield of an 100 keV-thick heavy ice target.

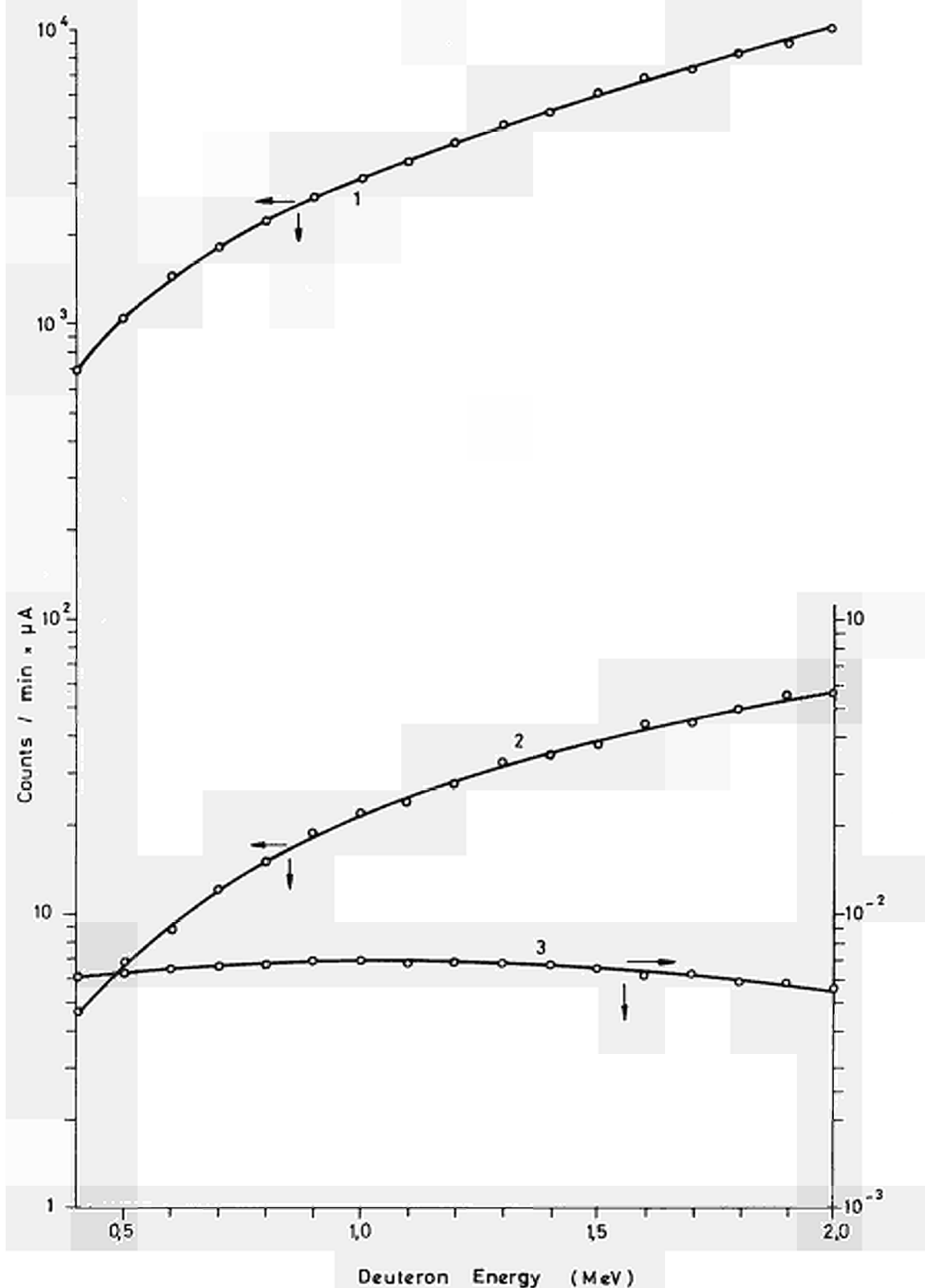


Figure 1.- Neutron yield from a heavy ice target and from the copper backing. Curve 1: number of counts for a 100 keV-thick heavy ice target. Curve 2: number of counts from the copper support for a clean, new target backing, obtained in the same experimental conditions as curve 1. Curve 3: ratio of curve 1 to curve 2.

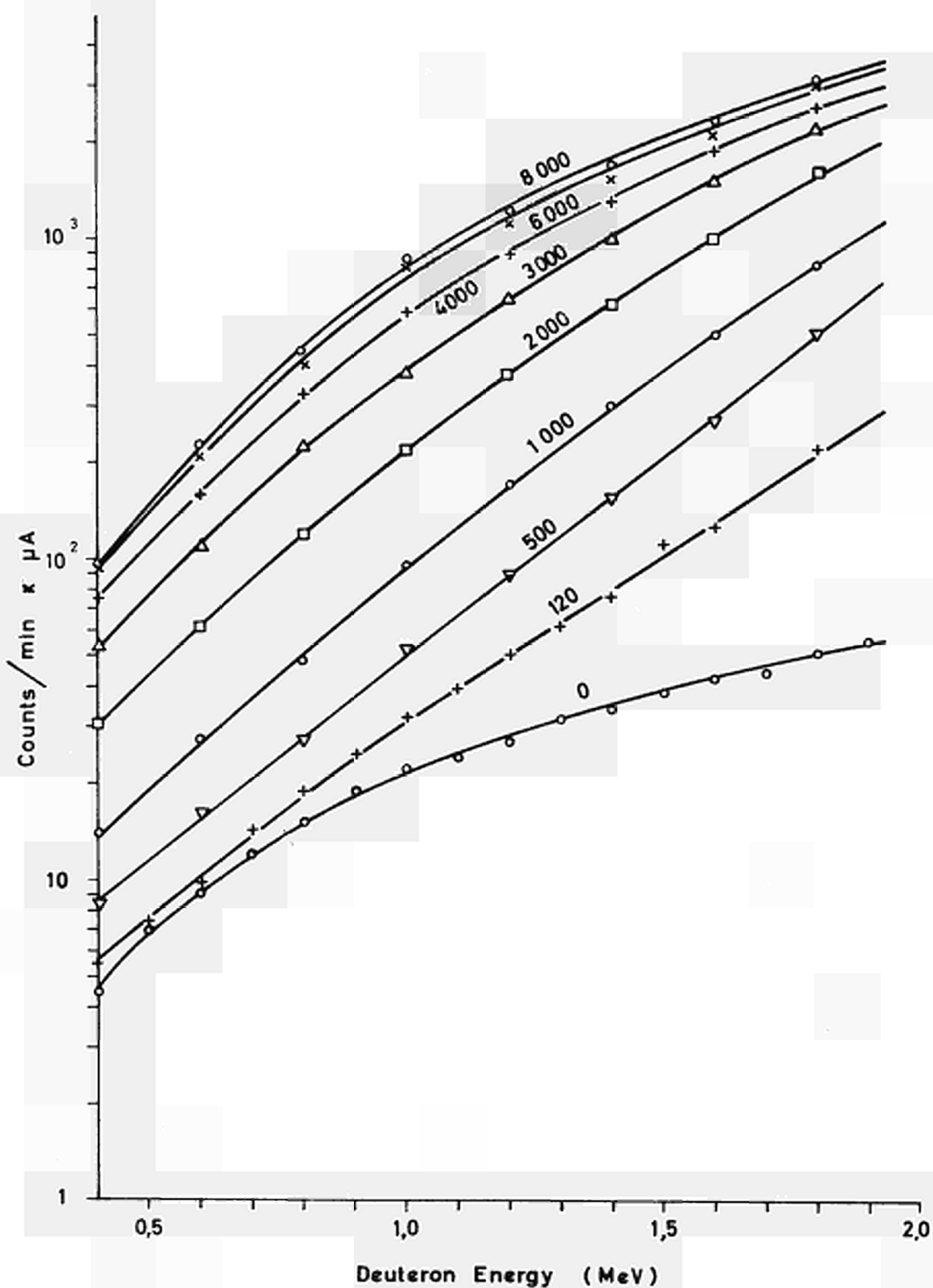


Figure 2.- Number of neutrons from the self-formed deuterium target vs energy, after several bombarding times.- Time is given in minutes.

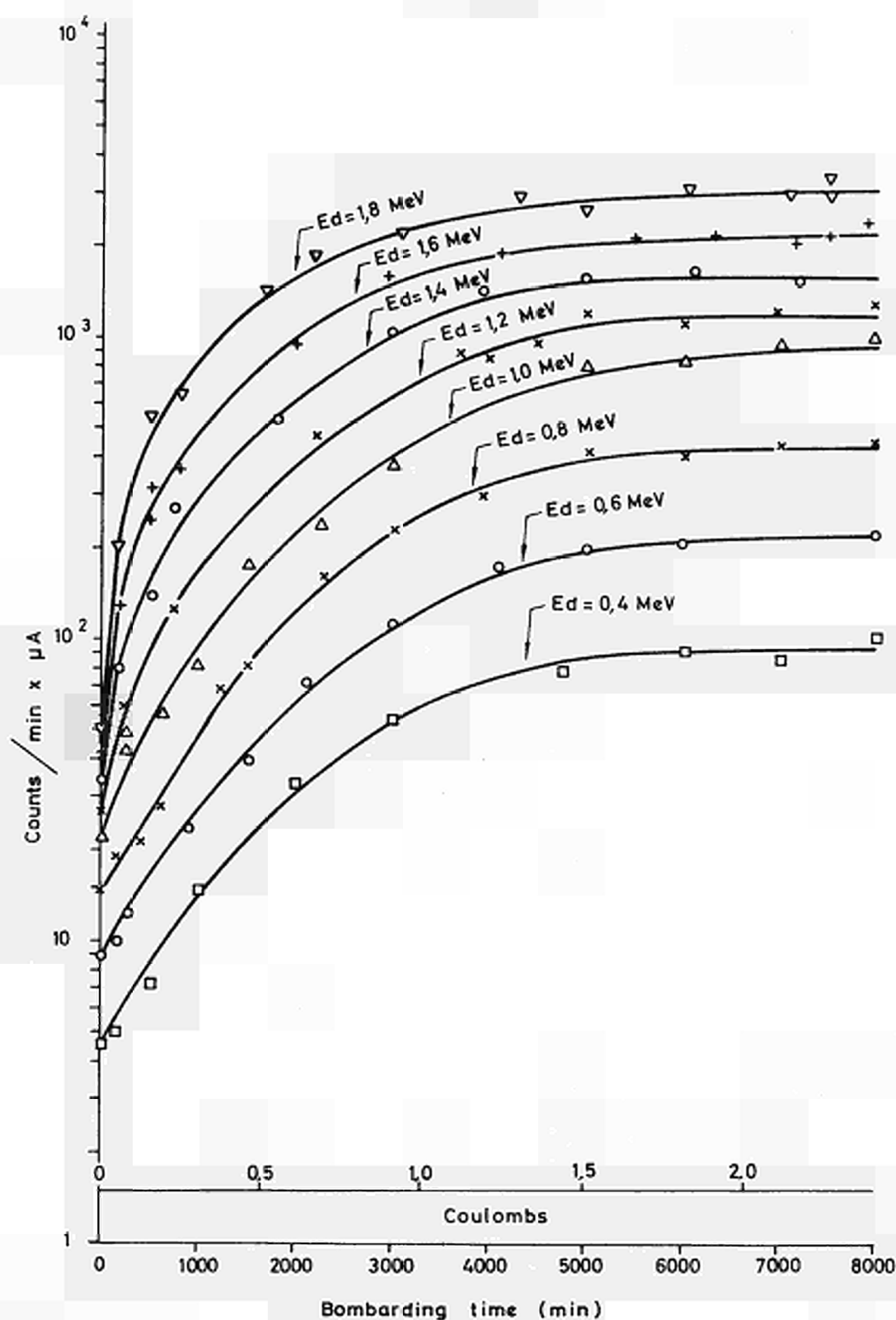


Figure 3.- Number of neutrons from the self-formed deuterium target vs bombarding time and deposited charge.

As it was already noted the target support was bombarded with deuterons of energies from 0,4 to 2,0 MeV. The deuterium will extend then to a depth corresponding to the maximum range of a 2.0 MeV deuteron, but with an unknown density distribution, that we can expect will be greater close to the surface, due to the increasing bombarding time with decreasing energy. Therefore the neutron yield *vs* energy and time should depend slightly on the bombardment history but the shapes of the curves and their saturation values should be very similar.

The self-target can be eliminated almost completely by sandpapering and polishing the target backing. It is interesting to notice as shows fig. 4, that a large contribution to the neutron background is given by the thin layer of carbon formed on the target support. In fact this graph shows that after removing the carbon film the neutron background decreases to a half of the total value. A smooth sand-papering of the backing can reduce the background in an order of magnitude, approaching the value of a clean new backing.

All these results show that the neutrons produced at the self-target can give an important contribution to the total number of counts registered at the detectors, and in those cases where they must be avoided, the target backing ought to be cleaned or changed regularly, or the neutron energy selected with the appropriate technics.

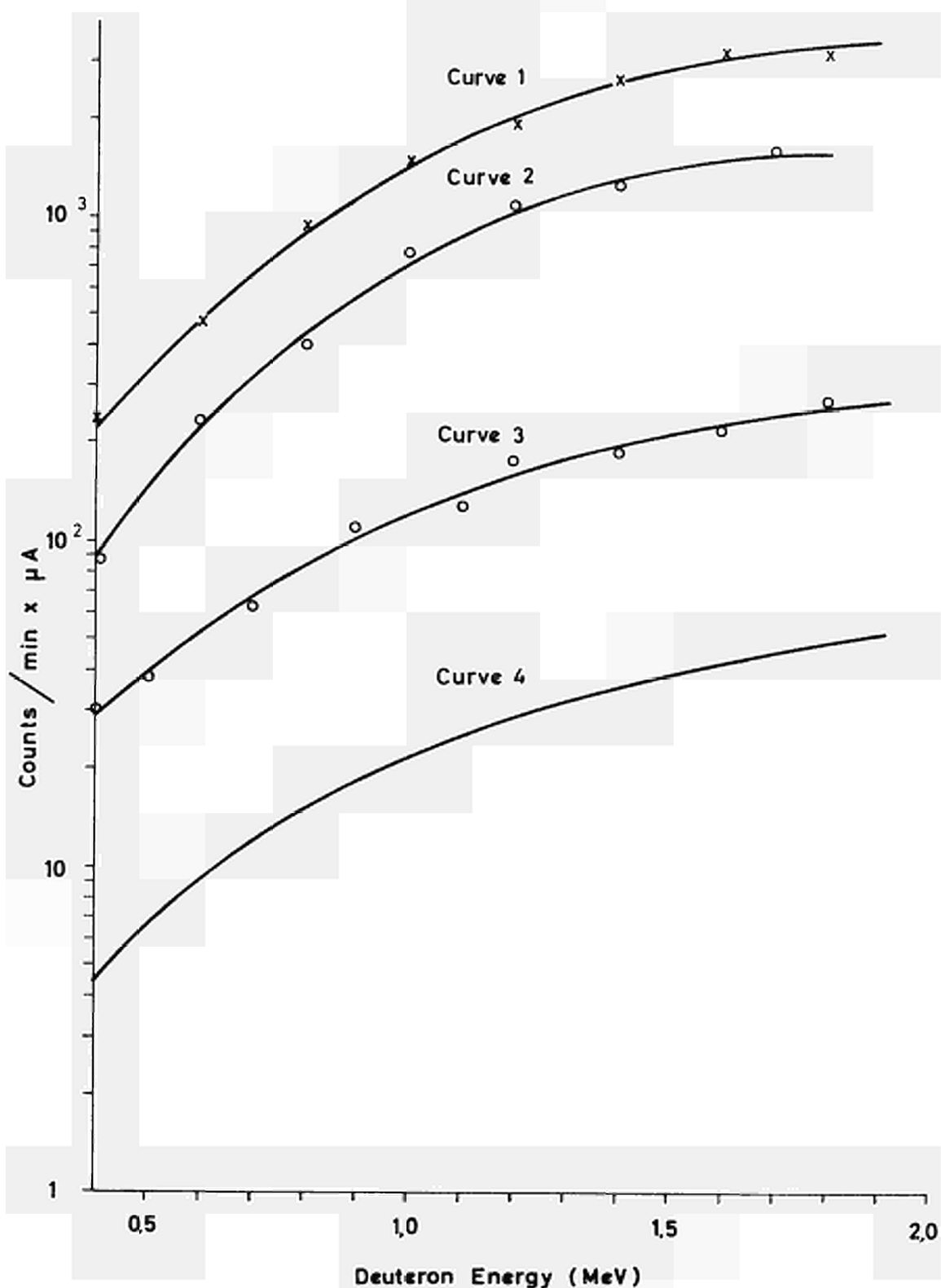


Figure 4.- Neutron yield from a used target backing. Curve 1: neutron background for a long bombarded target backing. Curve 2: Number of neutrons from the backing after removing the carbon deposit. Curve 3: Background after sandpapering and polishing the target backing. Curve 4: Background from a new, not bombarded target support.

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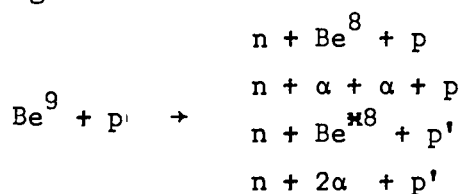
Beryllium Target for Neutron Production by $\text{Be}^9(\text{pn})$ reactions.

F. BODART and G. DECONNINCK
Université de Louvain, Belgique.

ABSTRACT

The $\text{Be}^9(\text{pn})$ reaction has been studied between 2.5 and 4 MeV incident energy by a time of flight method. The different groups of neutrons have been separated and the cross sections measured at different angles and energies. The most important group of neutrons emerges from the $\text{Be}^9(\text{pn})\text{B}^9$ reaction which can be utilised as a source of monoenergetic neutrons in some conditions. The advantages and possibilities of a Beryllium target are discussed.

$T(pn)He^3$ and $Li^7(pn)B^7$ reactions are the most utilised sources of MeV neutrons although some limitations due to target problems are often encountered. A much less known source of neutrons is the reaction of protons on Beryllium, it produces a group of monoenergetic neutrons from the $Be^9(pn)B^9$ reaction and a spectrum of "secondary" neutrons due to the following reactions



The aim of this work is to determine the number of secondary neutrons and to analyse the performances of $Be^9(pn)B^9$ as a source of monoenergetic neutrons. The preceding reactions have been carefully studied with good angular and energy resolutions, more than 60 spectra of neutrons have been detected at different angles between 2.6 and 4 MeV incident energy.

The advantages of utilising Beryllium as target are obvious : melting point, chemical stability, low cost, purity, good conductivity of heat and electricity ...

Apparatus.

The Van de Graaff accelerator of the University of Louvain is equipped with a 4 nsec pulsed source, a post acceleration electromagnetic deflector allows the production of 1,5 nsec burst of protons, the fig. 1 gives a schematic view of this assembly.

The Beryllium targets are easily produced by evaporation of Beryllium powder on aluminium backing, they have shown to be extremely stable under protons bombardment. Neutrons are detected by the time of flight technique with a basis of 200 cm, the detector itself is a stilbene scintillator (1" x 1") coupled on a 56 AVP photomultiplier, neutron pulses are selected by pulse shape discrimination.

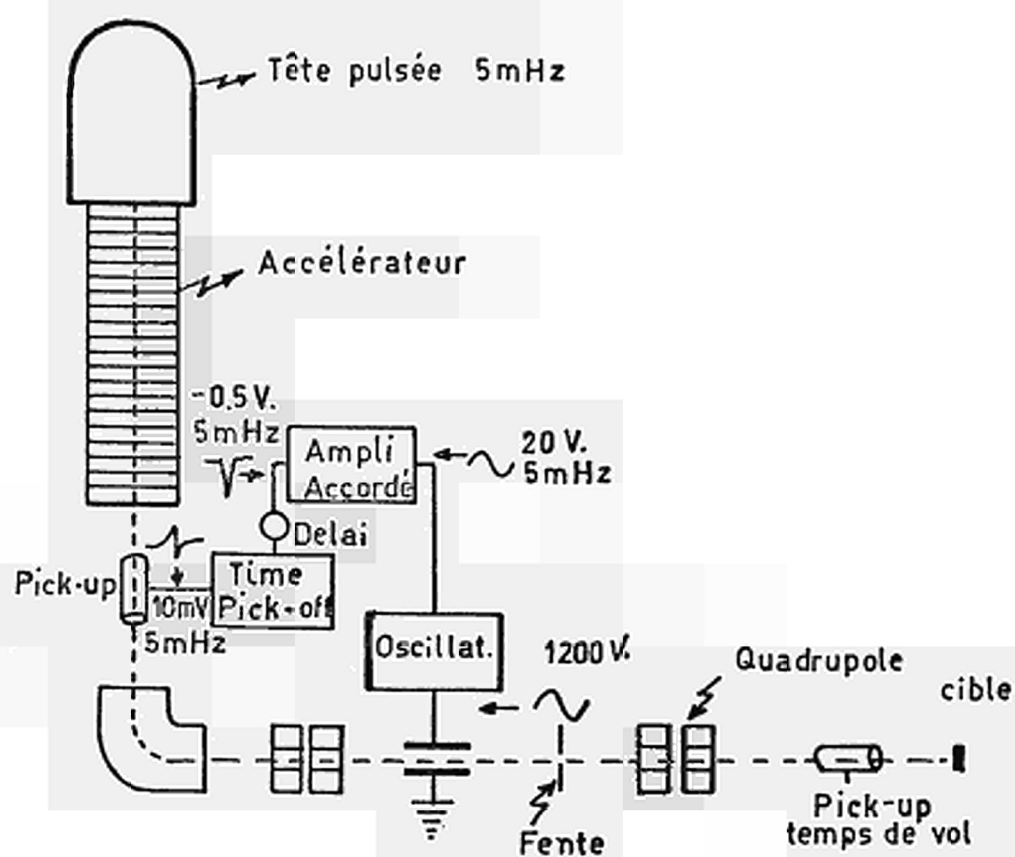
The detector is biased at a level corresponding to 700 keV and all the result presented have been obtained in this condition. The final resolution of the assembly varies between 1.5 and 3 nsec following work conditions. The efficiency of the detector has been measured with the neutrons of $T(pn)He^3$ and $Li^7(pn)B^7$ reactions.

Experimental results.

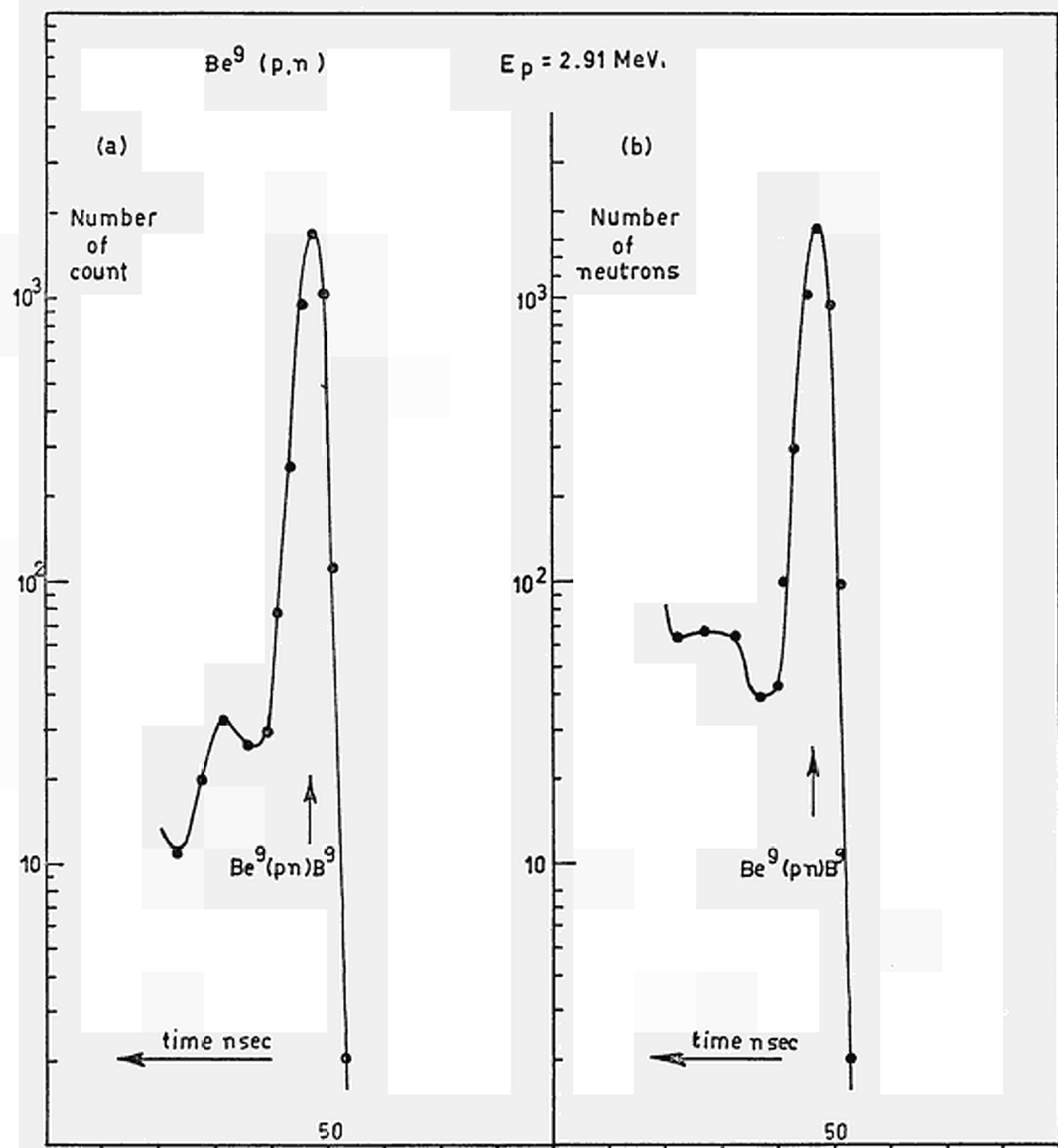
Fig. 2a gives an example of spectrum taken at 0° , in this figure the number of count effectively detected is plotted against the time of flight. There is a broad peak from $Be^9(pn)B^9$ neutrons and a background of other neutrons appears on the left of this peak, this spectrum ends suddenly at a point corresponding to 80 nsec, this cut is due to the 700 keV threshold. The relative importance of the $Be^9(pn)B^9$ reaction is clearly seen on this figure.

Fig. 2b shows the same spectrum corrected for efficiency and the curve represents the number of neutrons emitted at 0° .

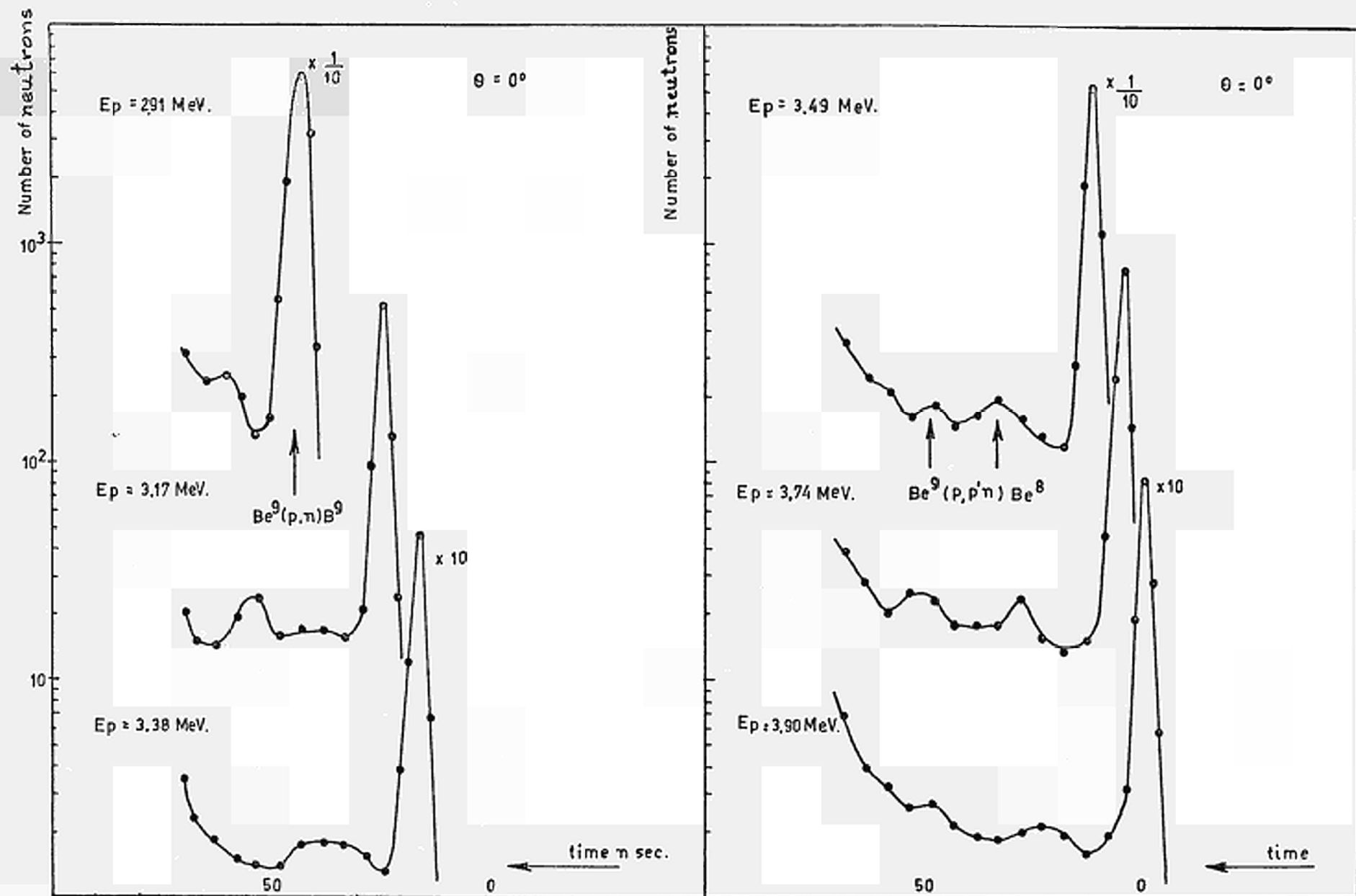
Fig. 3 is a sample of spectrum taken at 0° and corrected for efficiency, it is clearly seen on this picture that the background of secondary neutrons increases rapidly with the proton energy.



- fig. 1 - Equipement utilised for (pn) reactions measurements



-fig. 2 - (a) Number of count detected in a Stilbene cristal (Bias = 700 KeV.)
 (b) Curve (a) Corrected for efficiency of the detector



-fig. 3 - $\text{Be}^9(p,n)$ reactions - On example of angular distributions taken at 0° (flight path 200 cm)

Neutrons emitted at zero degree are the most important when this reaction is used at neutron source, for this reason a plot of the cross section of the $\text{Be}^9(\text{pn})\text{B}^9$ reaction is given in fig.4. The errors are mainly due to uncertainty in the calibration, the point at 3.06 MeV has been used as normalisation point by comparison with the $\text{Li}^7(\text{pn})\text{B}^7$ reaction, an additional error of 10 % can be estimated as a result of this procedure.

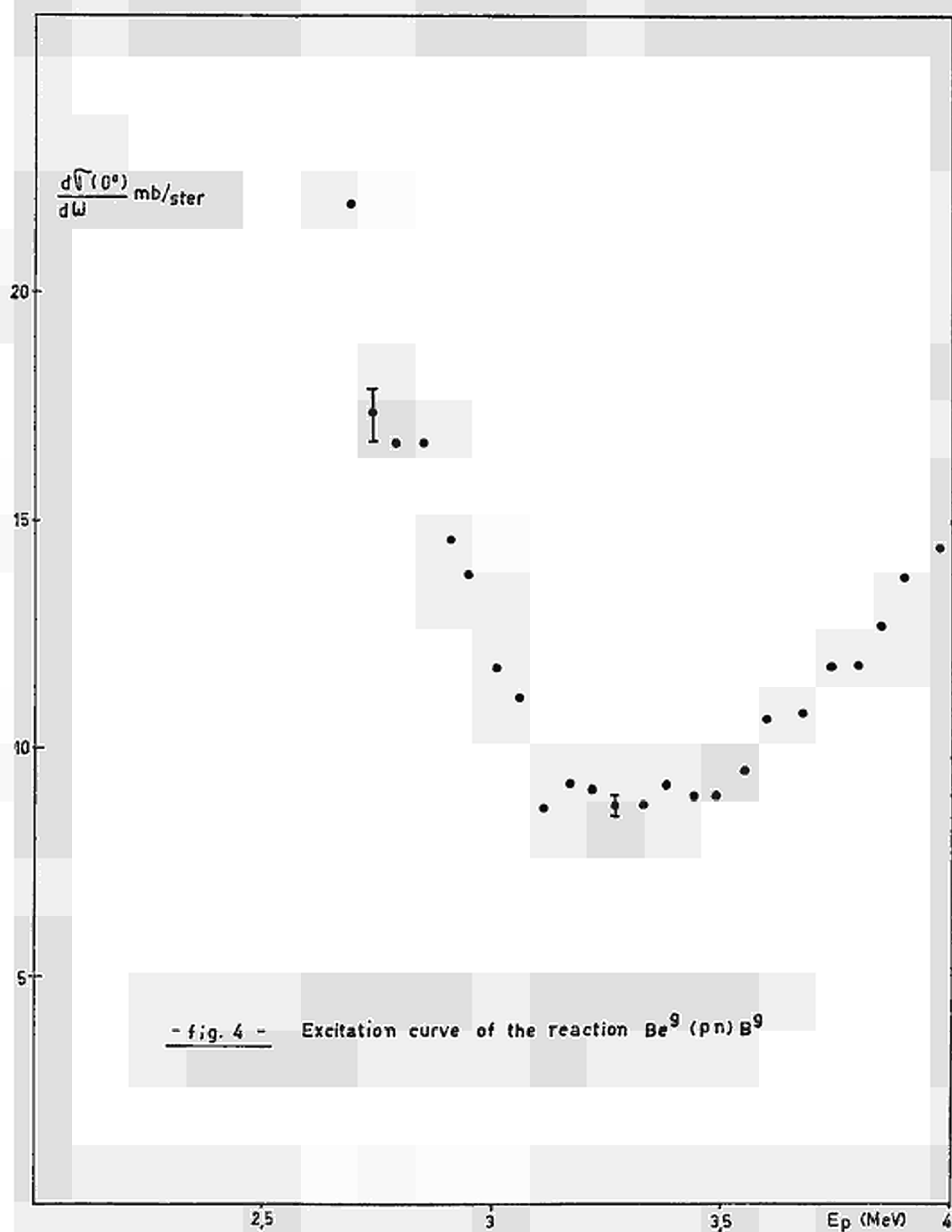
Table I gives an estimation of differential cross sections of neutrons of different energies, the background has been integrated on 100 keV intervals and the $\text{Be}^9(\text{pn})\text{B}^9$ cross section is shown in the last column. This table allows one to estimate the relative importance of secondary neutrons, an error of about 30 % can be estimated on the figures given in this table.

The $\text{Be}^9(\text{pn})\text{B}^9$ as a source of monoenergetic neutrons.

1. Monoenergetic neutrons.

- The reaction $\text{Be}^9(\text{pn})\text{B}^9$ has a threshold of 2.060 ± 0.0005 MeV and the neutrons emitted are thus monoenergetics, this threshold may be compared with the $\text{Li}^7(\text{pn})\text{B}^7$ thresholds (1.889 MeV).

- The importance of secondary neutrons is shown in table I but the number of really detected with a biased stilbene detector can be estimated from fig.2a, the ratio of "good" neutrons to background neutrons is less than 0.05 at 3 MeV, but goes up to 0.30 at 4 MeV. It is to remember that the $\text{Li}^7(\text{pn})$ reaction has also a background of secondary neutrons in this energy region. If the detection is made by the time of flight technique, these figures goes down to 1% at 3 MeV and a few percent at 4 MeV (better ratios can be obtained with longer flight path). If the detection is made with a long counter, the neutrons detected can not be considered as monoenergetic (see table I).



- fig. 4 - Excitation curve of the reaction $\text{Be}^9(p,n)\text{B}^9$

E_n MeV E_p MeV	.75	.85	.95	1.05	1.15	1.25	1.35	1.45	1.55	1.65	1.75	1.85	1.95	2.05	2.15	2.25	$\frac{d\sigma}{d\omega} (0^\circ)$ $Be^9(pn)B^9$
2.69																	21.91
2.74	(1.7)																17.34
2.79	(1.7)	0.1															16.70
2.85	(1.7)	0.2															16.70
2.91	(1.8)	0.8	0.5														14.57
2.95	(1.8)	0.9	0.6	0.4													13.84
3.01	(2.6)	0.8	0.9	0.9	0												11.77
3.06	(2.3)	1.0	0.9	0.9	0.2												11.1
3.11	(1.4)	1.1	0.7	0.6	0.5												8.68
3.17	(1.1)	1.3	0.7	0.6	0.5	0.5											9.26
3.22	(1.1)	1.0	0.9	0.7	0.5	0.5	0.3										9.11
3.27	(1.1)	0.8	0.9	0.7	0.6	0.4	0.3										8.74
3.33	(1.1)	0.8	0.8	0.8	0.6	0.4	0.3	0.1									8.80
3.38	(1.7)	1.0	0.8	0.8	0.7	0.5	0.3	0.2									9.20
3.44	(1.7)	1.0	0.9	0.8	0.7	0.4	0.3	0.2	0.2								8.98
3.49	(2.1)	1.1	0.8	0.8	0.7	0.5	0.3	0.2	0.2								9.00
3.55	(2.3)	1.0	0.8	0.8	0.7	0.5	0.3	0.3	0.3	0.1							9.52
3.62	(2.1)	1.1	0.9	0.9	0.7	0.6	0.4	0.3	0.3	0.2							10.66
3.68	(2.1)	0.9	0.8	0.8	0.7	0.7	0.4	0.3	0.3	0.2	0.1						10.70
3.74	(2.1)	1.5	1.0	0.9	0.7	0.8	0.4	0.4	0.3	0.3	0.2	0.2					11.75
3.80	(3.2)	1.5	1.0	0.9	0.7	0.8	0.6	0.4	0.3	0.3	0.3	0.3	0.1				11.82
3.85	(4.0)	1.8	1.2	1.0	0.7	0.7	0.6	0.4	0.4	0.4	0.3	0.4	0.2				12.67
3.90	(4.1)	1.8	1.2	0.9	0.7	0.6	0.7	0.5	0.4	0.4	0.4	0.4	0.4	0.1	0.1		13.79
3.98	(4.9)	2	1.2	1.0	0.7	0.7	0.6	0.5	0.4	0.4	0.4	0.5	0.4	0.2	0.2	0.1	14.44

- The ratio between $\text{Be}^9(\text{pn})\text{B}^9$ and $\text{Li}^7(\text{pn})\text{Be}^7$ cross sections is about $\frac{1}{4}$ at 3 MeV proton energy and better elsewhere, this source of neutrons is thus intense enough for MeV neutrons experiments. We must finally point out that the variation of energy of emitted neutrons with angle is smaller in the case of Be than in the case of Li, this can be an advantage in some experiments (fig.5).

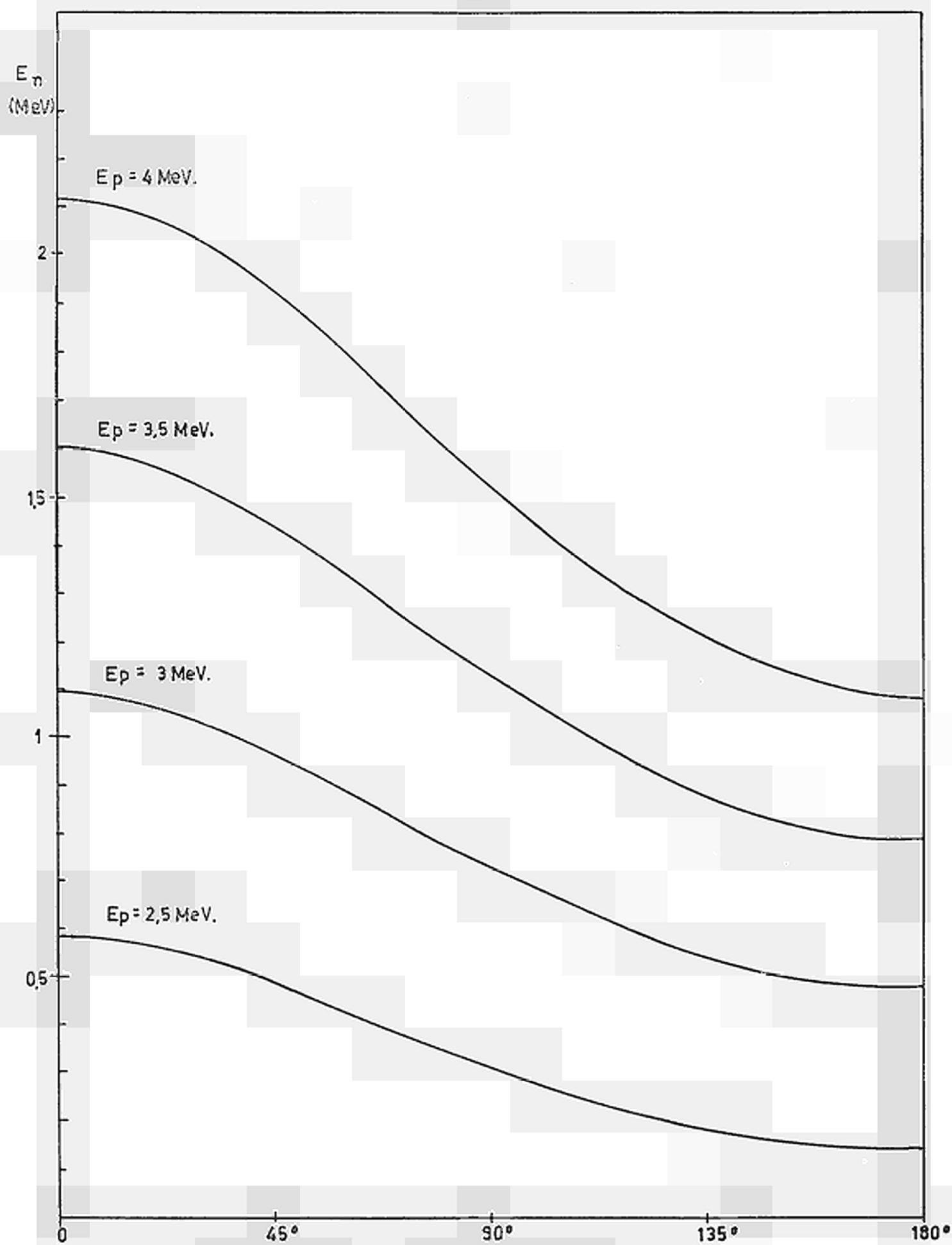
- For all these reasons, we can consider that the $\text{Be}(\text{pn})$ reaction can be utilised as a source of monoenergetic neutrons between 2.5 and 4 MeV, if the detection is made by biased or better by time of flight, the use of long counter is to be avoided.

2. Beryllium as a target.

Beryllium has been long used in cyclotron for the production of neutrons by $\text{Be}^9(\text{dn})$ reactions, the main advantages of this substance is the high melting point, good conductivity of heat and electrical charges, this allows the use of very intense beams.

Self-supported Beryllium target are easily made by evaporation and very thin target can be deposited on metal backing for high resolution experiment and the handling of these targets is extremely easy in open air as well as in vacuum (low chemical reactivity).

The thickness of the target can be measured by the threshold method (detection of thermalised neutrons emitted at 0°) with high accuracy; with a Van de Graaff giving protons of 2.000 ± 0.001 MeV an accuracy of 1% is obtained for a 1 mg/cm^2 target.



- fig. 5 - Neutrons from the $\text{Be}^9(p,n)\text{B}^9$ reaction

DISCUSSION

Mr. MORGANSTERN

What is the pulse amplitude ? At what distance ?

Mr. DECONNINCK

The average proton intensity was 1 μ A during the measurements, but this is only a limitation of the time of flight and of the type of source used.

The distance was 200 cm, with a longer distance the proportion of background neutrons can be reduced to much less than 1%.

Mr. FLEISCHER

What is neutron yield per microamp ?

What is neutron energy ?

Mr. DECONNINCK

About 10^7 n/ μ coulomb, steradian. The energy of the neutron is indicated on fig. 5.

Mr. PAIC

Quel est l'intérêt de la réaction Be^9 (p,n) puisque son intensité est inférieure à celle de la réaction Li^7 (p,n)

Mr. DECONNINCK

J'ai cité les avantages au cours de l'exposé qui précède, mais je peux résumer en disant que l'avantage du beryllium est sa nature métallique qui lui permet de recevoir des faisceaux très intenses de protons ce qui n'est pas le cas du lithium ou de ses composés (LiF , Li_3N , Li_2O), de plus la composition des cibles de beryllium est toujours homogène.

Mr. PEISACH

Could you tell us what detector you used.

Mr. DECONNINCK

Stilbene

Mr. PEISACH

The pulse shape discrimination enabled you to cut out neutrons below 700 keV. Your neutron energy measured with 3 MeV protons was about 1 MeV. What was the energy of your background ?

Mr. DECONNINCK

Continuous background for 700 keV to the bombarding energy.

DEMONSTRATION NEUERER TARGETENTWICKLUNGEN

H. FABIAN

NUKEM GmbH Wolfgang b/Hanau-Allemagne

Zunächst muss ich dem Herrn Vorsitzenden danken, dass er mir erlaubt hat, ausser Programm Ihnen, meine Damen und Herren, unsere Targettypen und Konstruktionen zu zeigen.

Es ist für den Experimentator wichtig, glaube ich, bei der Vorbereitung von Experimenten zu wissen, welche Möglichkeiten in Bezug auf Targets gegeben sind, d.h. welche Targetmaterialien, welche Formen und Dimensionen und welche Konstruktionen zur Verfügung stehen.

In Abb. 1 sind unsere Standardabmessungen auf 0,3 mm Cu-Unterlage zu sehen; von 0,5" bis 1 13/16" voll bedampft. Da beim Einspannen der Rand des Targets nicht ausgenutzt wird, ist es ökonomischer, diesen Rand überhaupt nicht zu bedampfen. In Reihe 2 sind diese Abmessungen von 1" - 1 1/8" bis 1 9/16" - 1 13/16" abgebildet.

Abb. 2 zeigt diverse Unterlagen (Mo, Ta, W, Au, V2A, Al) mit Titan bedampft.

In Abb. 3 sind Targets aus Er, Y, Sc, La, Ce, Zr auf Mo-Unterlage zu sehen.

Freitragende Titanfolien von 0,5 μ oder mehr in Fassungen sind in Abb. 4 dargestellt.

Flansche und Konstruktionen (von beiden Seiten fotografiert) sind auf Bild 5 a bzw. 5 b zu sehen. Unser rot. Flansch mit 100 cm² effektiver Fläche und cirkularen Kühlrippen ist für besonders hohe therm. Belastung gedacht. Nach den Berechnungen von Herrn Cossuta von heute morgen, 330 W/cm² Wärmeabfuhr, wird unser Flansch 33 kW ableiten können. Dies entspricht einer Neutronenausbeute von $1,3 \cdot 10^{13}$ n/sec, extrapoliert von unseren Ausbeuten bei 2 mA und

250 kV mit $2 \cdot 10^{11}$ n/sec. Um die Ausbeute und Lebensdauer solcher Targets zu messen hätten wir gerne einen entsprechenden Beschleuniger.

Für die Verwendung in einer Maschine mit der Neutronenausbeute von $5 \cdot 10^{13}$ n/sec bei 800 mm Strahldurchmesser ist eine Konstruktion entwickelt worden, um die Targetfläche aus einzelnen Segmenten zusammensetzen zu können. Dies bringt Vorteile bei der Herstellung und Imprägnierung der Titanschichten, beim Umgang mit Tritium und ermöglicht ausgeschossene Segmente zu vertauschen bzw. auszutauschen.

Wir glauben durch die Entwicklung von rot. Targets und Grossflächen-targets die durch leistungsschwache Targets bedingte Grenze im Beschleunigerbau abgetragen zu haben und hoffen nur, dass uns Maschinen mit einigen 100 mA bald zur Verfügung stehen werden.

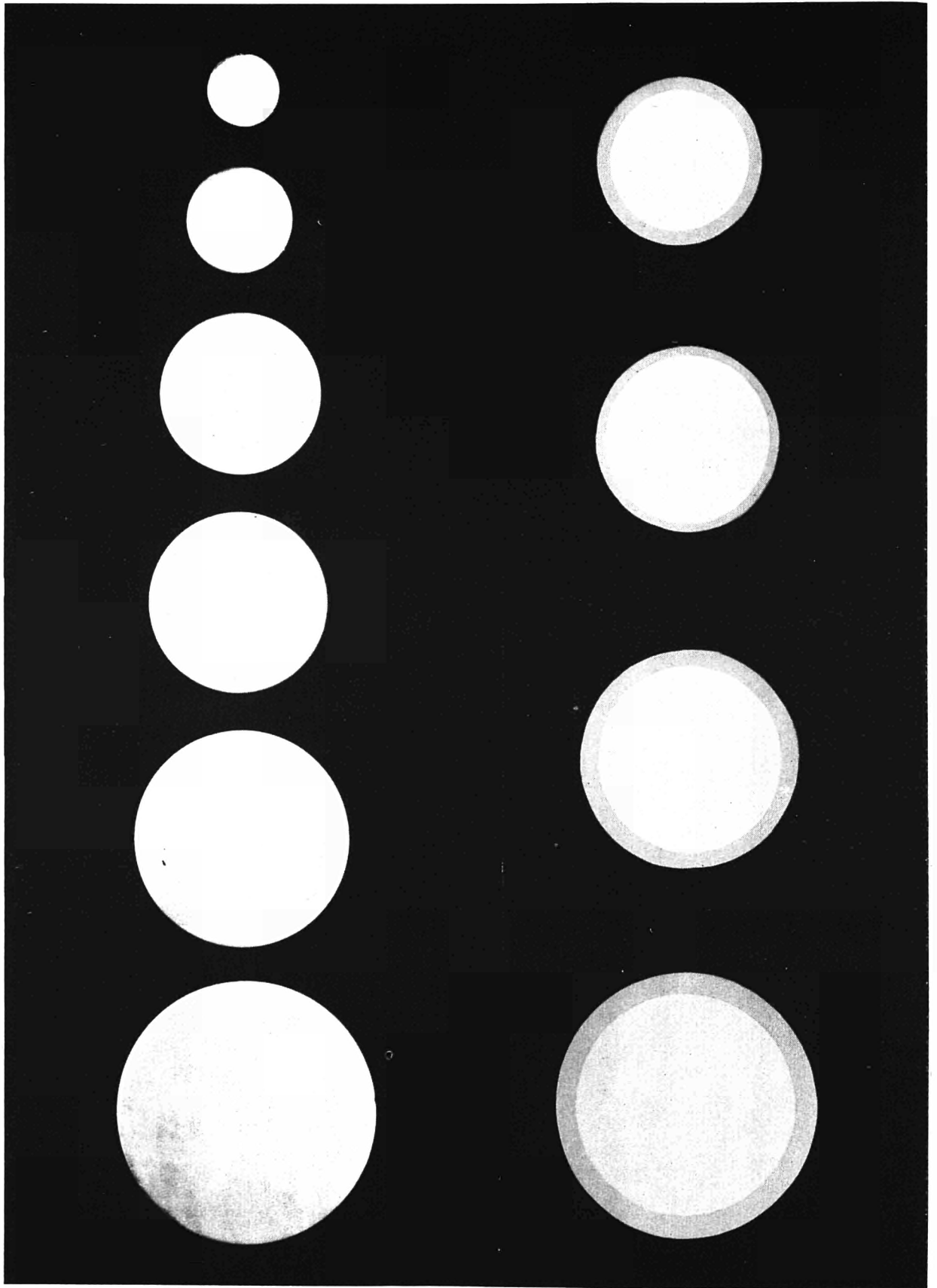


Abb. 1a

Abb. 1b

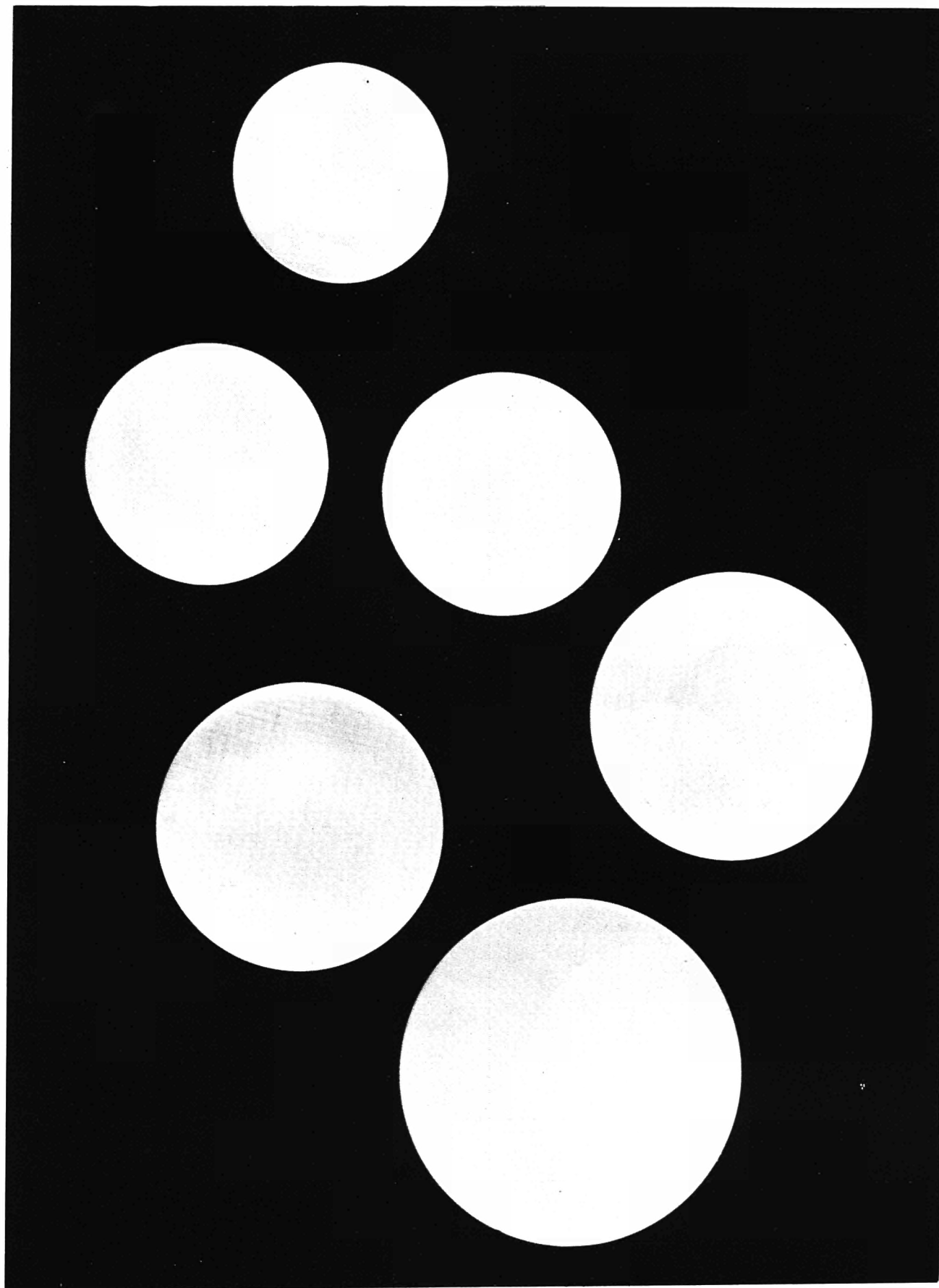


Abb. 2

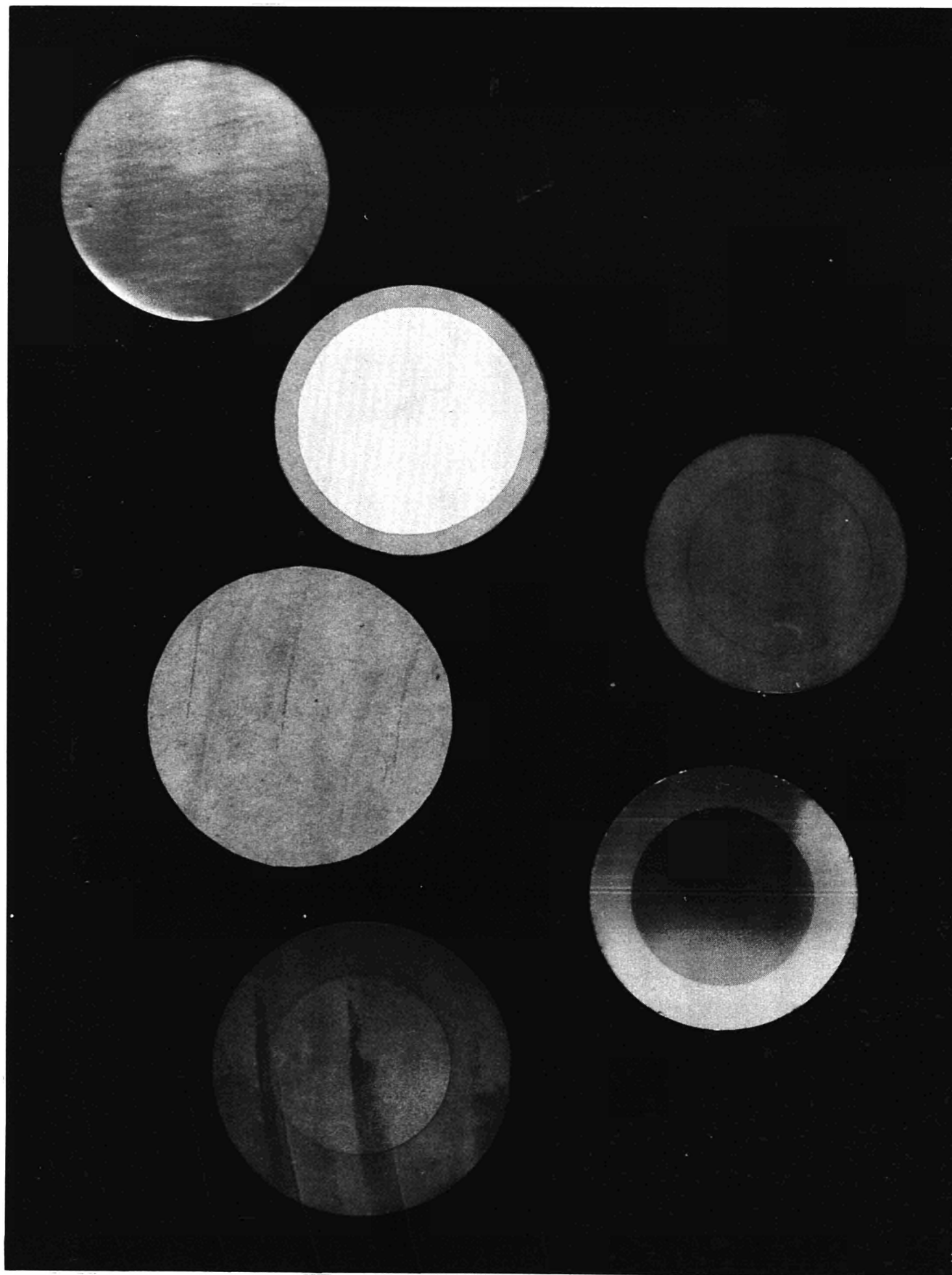


Abb. 3

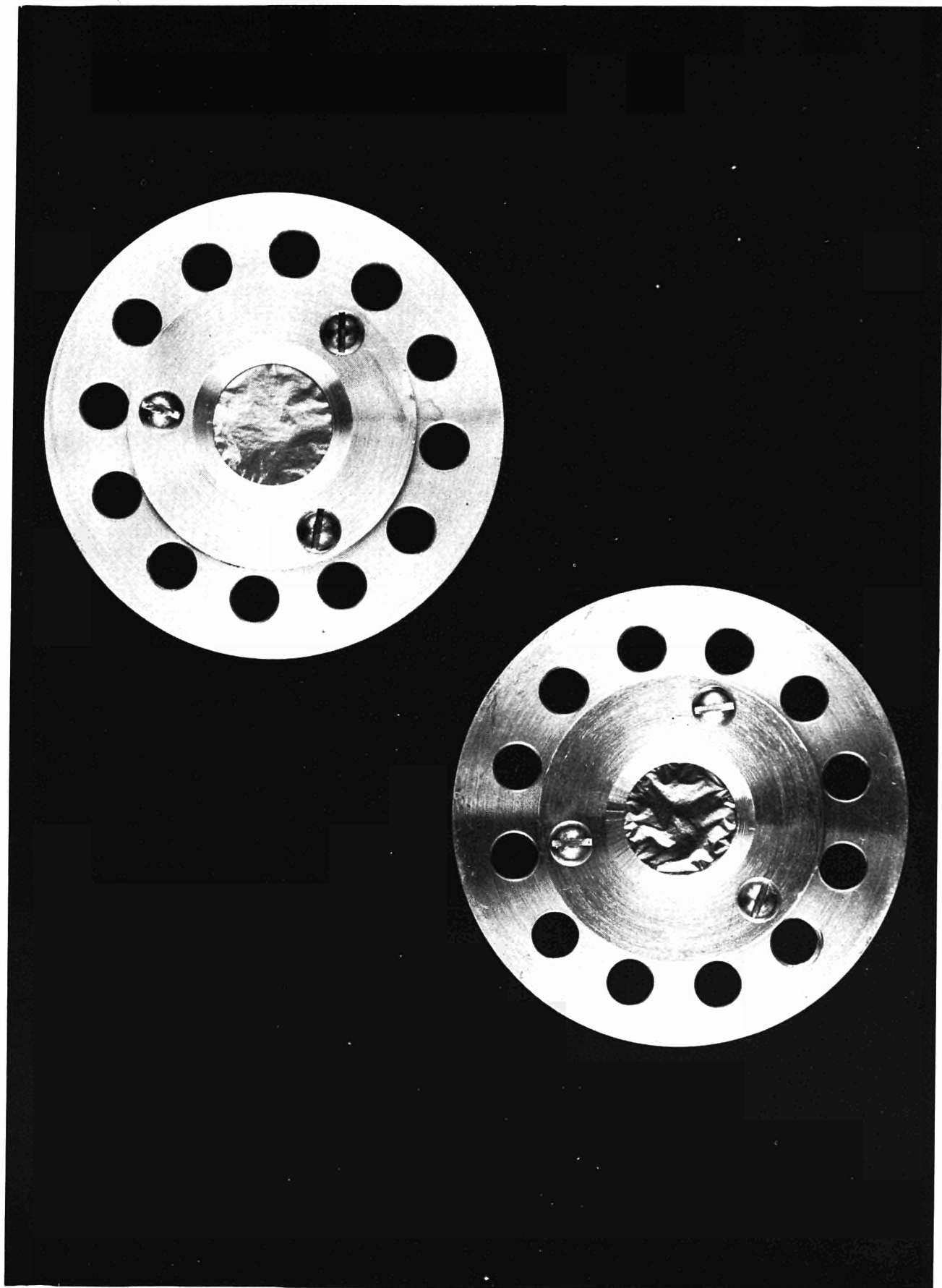


Abb. 4



Abb. 5a

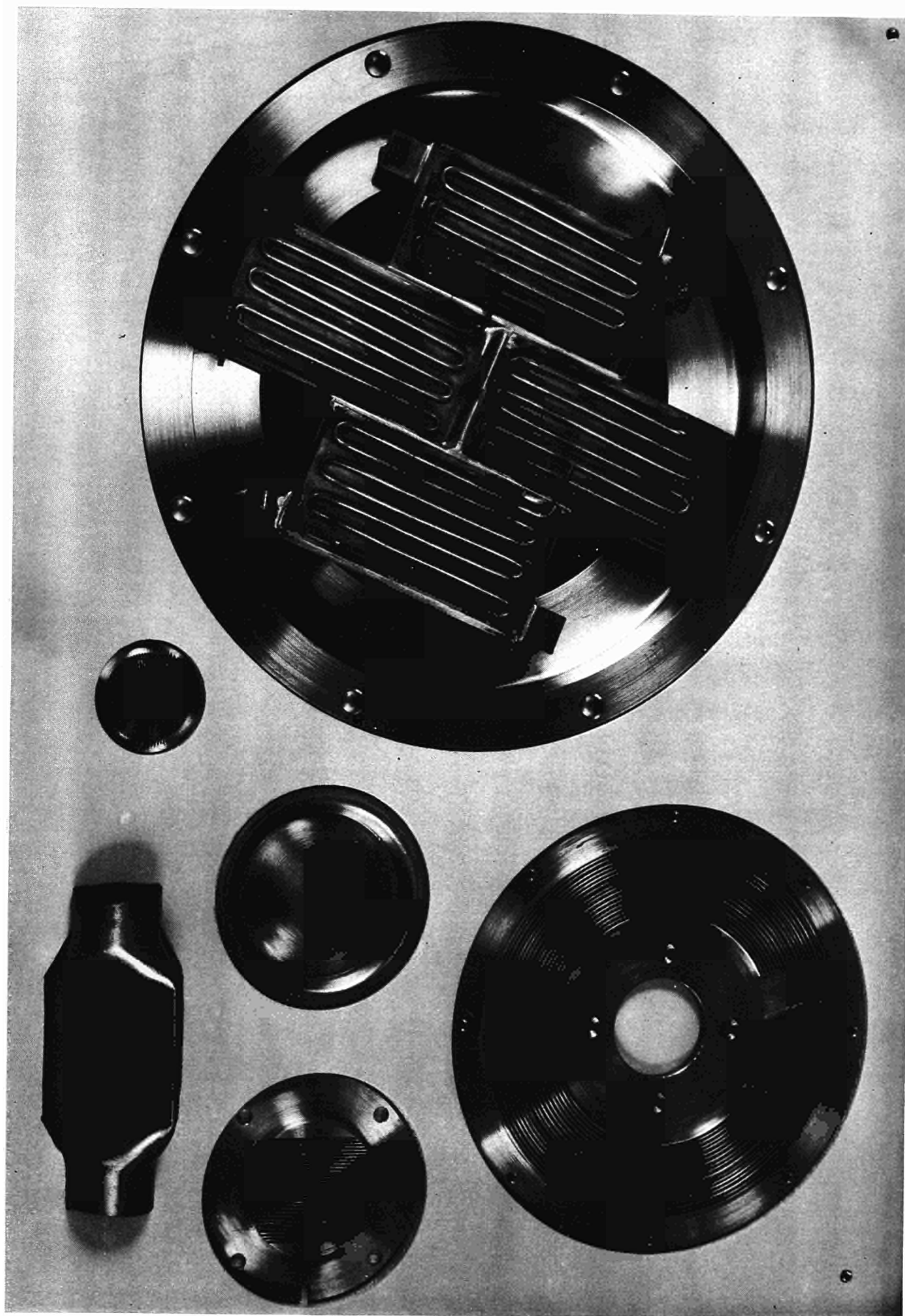


Abb. 5b

THE PRODUCTION OF FAST NEUTRONS BY He^3 INDUCED
NUCLEAR REACTIONS

E. Bruninx and J. Crombeen
Philips Research Laboratories
N.V. Philips' Gloeilampenfabrieken
Eindhoven Netherlands

ABSTRACT

The energy spectrum of the fast neutrons produced in the forward direction by 20 MeV He^3 particles on a beryllium target has been measured using nuclear reactions with different reaction thresholds. The results were interpreted by means of an empirical formula. At the same time the angular distribution of the fast neutrons was determined and the total yield/microcoulomb estimated.

I. Introduction

Various methods are known for the measurement of fast neutron spectra. Proton recoil techniques, scintillators, solid state detectors have been used for this purpose ¹⁾. Although they are able to give good results the amount of effort required is rather large.

By far the simplest method for measuring fast neutron spectra is based upon the use of nuclear reactions (activations) with different reaction thresholds. This method of employing various detector foils is probably the oldest one, already used many years ago in the determination of reactor neutron spectra ²⁾. The measured fission neutron spectra could be described (represented) by a simple empirical formula.

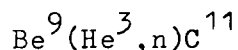
Recently an extension of this empirical formula has been proposed by Heertje et.al. ³⁾ who measured fast neutron spectra produced by 26 MeV deuterons on various thick targets.

For the measurement of the absolute number of neutrons produced the technique of the manganese bath has been mainly employed ¹⁾. It is liable to give good results but the set-up is quite complicated.

Using the above mentioned method of nuclear activation it is however equally feasible to calculate in a simple way the flux at the position of the detector foils.

By measuring the angular distribution of the fast neutrons we can integrate the flux over all angles and obtain the total yield/micro-coulomb.

By the afore mentioned technique we measured the energy spectrum, the angular distribution and the total production rate of the fast neutrons for the reaction



II. Principle of the method

Foils of Aluminium, Silicon, Copper and Carbon (as a plexiglass disc) are stacked in a sandwich and irradiated in the fast neutron beam at some distance from the beryllium target for a given period of time, so as to induce a sufficiently well measurable activity. Following the irradiation, absolute disintegration rates of the reaction products (Mg^{27} , Na^{24} etc...) are determined using calibrated counters (see further section on Experimental techniques). During the bombardment the intensity of the He^3 beam is continuously measured.

The nuclear properties of the various detector foils are summarized in table I.

Table I

Nuclear Reaction	E(threshold) MeV	Reference for cross-section
$\text{Al}^{27} \text{ (n.p) } \text{Mg}^{27}$	3.00	4.5
$\text{Si}^{28} \text{ (n.p) } \text{Al}^{28}$	4.00	6,7
$\text{Al}^{27} \text{ (n,}\alpha\text{) } \text{Na}^{24}$	6.00	8
$\text{Cu}^{63} \text{ (n,2n)Cu}^{62}$	11.5	9, 10
$\text{C}^{12} \text{ (n,2n)C}^{11}$	20.0	11

At higher energies not all cross sections were known. The values at lower energies were then simply extrapolated. The reaction threshold as given above indicates only the value at which the cross section is measurable (i.e. > 1 mb); it does not represent the true "physical" or "theoretical" threshold. The thresholds of the various reactions cover the whole energy spectrum of the produced fast neutrons. At least 3 different reactions are required, but the more detectors the better the definition of the spectrum shape.

The basic assumption in the interpretation of the induced absolute disintegration rates of the different threshold detectors is that the shape of the neutron spectrum can be represented by a formula

$$N(E) = K \cdot E^n \cdot \exp - \frac{E}{\xi} \quad (1)$$

where $N(E)$ = number of neutrons of a given energy

K = constant

n, ξ = parameters determining the shape and mean energy of the neutron spectrum.

Fission neutron spectra were fitted to a similar expression with values of $n = 0,5$ and $\xi = 1.29$ ¹²⁾.

There is no direct physical or mathematical proof for this formula. However the compound nucleus model, based upon variables such as excitation energy, can account by a similar formula, for the spectrum of neutrons evaporated from a nucleus excited to several tens of MeV. ¹³⁾

The total absolute disintegration rate for a given nucleide e.g. Cu^{62} formed by the reaction $\text{Cu}^{63} (n, 2n) \text{Cu}^{62}$ is

$$A = M_T \cdot N \cdot \langle \sigma \rangle \quad (2)$$

where A = total absolute disintegration rate

M_T = number of target atoms

N = neutron flux

$\langle \sigma \rangle$ = average cross section for the considered nuclear reaction (i.e. averaged over all energies for the fast neutrons produced above the threshold).

Expression (2) can be written somewhat more explicitly as

$$A = M_T \cdot \int_0^{\infty} N(E) \cdot \sigma(E) \cdot dE \quad (3)$$

where $\sigma(E)$ cross section at a well defined neutron energy.

For each one of the threshold detectors used a similar expression holds.

Introducing now (1) into (3) yields.

$$A = M_T K \int_0^{\infty} \sigma(E) E^n \exp - \frac{E}{\xi} dE \quad (4)$$

or

$$A = M_T \cdot K \cdot S \quad (4a)$$

We have now a set of 4 such equations (corresponding to e.g. 4 threshold detectors used). In these equations A , M_T and $\sigma(E)$ are known; n and ξ are the two parameters whose value has to be determined. This is done by

assuming some arbitrary value of n and ξ and substituting into (4). One calculates now the values A_1/A_1 ($\equiv 1$), A_2/A_1 , A_3/A_1 etc..... and also similarly $(M_T.K.S)_2/(M_T.K.S)_1$ etc.....

and adjusts the values of n and ξ until the ratios of the A's and the $(M_T.K.S.)$'s correspond.

The absolute flux (at the position of the sandwich detector) can be obtained by integrating eq.(1) over all neutron energies

$$\text{thus} \quad \int_0^{\infty} N(E).dE = K. \int_0^{\infty} E^n \exp - \frac{E}{\xi} .dE \quad (5)$$

and substituting (4) \rightarrow (5) and rearranging we obtain

$$\varphi = \int_0^{\infty} N(E).dE = \frac{A}{M_T} \frac{\int_0^{\infty} E^n \exp - \frac{E}{\xi} .dE}{\int_0^{\infty} \sigma(E).E^n \exp - \frac{E}{\xi} .dE} \quad (6)$$

III. Experimental techniques

The experimental lay out is shown in fig. (1). The beam is extracted from the AVF prototype cyclotron and after focussing and deflection directed onto the beryllium target at the bottom end of the Faraday cup. The entrance of the Faraday cup contains 2 tantalum diaphragms shielded from the rest of the cup, the latter being equally isolated electrically so as to measure the total amount of current falling onto the target. Beam currents usually ranging from 2-15 μ A are measured via a 1M Ω resistor with a vacuum tube voltmeter, and registered as a function of time.

The detector foils consist of disc of 18, 10 or 5 m/m diameter and are placed at distances of 4.75, 7 or 10 cm from the target at 0°.

After activation the detector foils are counted with either a calibrated 3 x 3" NaI (Tl) crystal coupled to a TMC multichannel analyser or with a calibrated γ - γ coincidence counter.

Table II summarises the country techniques used together with the adopted half lifes.

fig.1

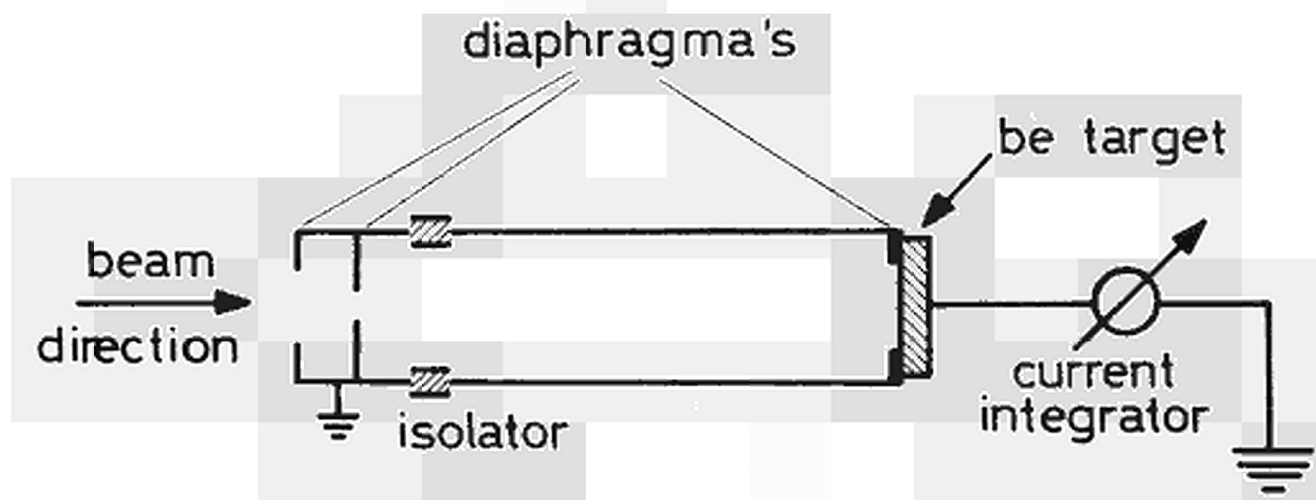


Table II

Reaction	t/2	Counting technique
Al $^{27}(n,p)Mg^{27}$	9.5'	γ , NaI (Tl) (3x3")
Al $^{27}(n,\alpha)Na^{24}$	15h	" "
Si $^{28}(n,p)Al^{28}$	2.3'	" "
Cu $^{63}(n,2n)Cu^{62}$	9.8'	β^+ coinc.
C $^{12}(n,2n)C^{11}$	20.3'	β^+ NaI (Tl) (3x3")

The counting rates were then corrected for efficiency, saturation factor, branching ratio, beam current etc.... and finally expressed as disintegrations/min (or sec)/ microcoulomb.

These values together with the values of M_T , the cross section σ and a rough estimate of n and ξ were then used to calculate the optimal values of n and ξ , and the total flux. A small computer programme was used for this purpose in order to reduce the tedious calculation by hand.

Angular distributions were measured simply by putting foils equidistant from the target at different angles around the target. After activation only relative measurements were taken i.e. the knowledge of the absolute disintegration rate is not required since all yields or counting rates are always normalized to 0° values.

IV. Results

- A) Sandwiches of the detector foils were irradiated at 0.9, 4.75 and 10 cm distance at 0° angle. Virtually all irradiations were performed at 20 MeV, but in order to test the method a few runs were made at lower and higher bombarding energy.

The results in fig.(2) represent neutron spectra at 14.7 and 27 MeV He^3 bombarding energy, taken at a distance of 0.9 cm from the Beryllium target. The optimized values of n and ξ were substituted in (1), and the total surface under the curve normalized to 1.

There is a gradual shift -as one might expect- but raising the bombarding energy by a given amount does not imply an identical increase in mean energy of the neutrons.

The mean energy at larger distances is somewhat higher, mainly due to the smaller contribution of lower energy neutrons sent off at larger angles.

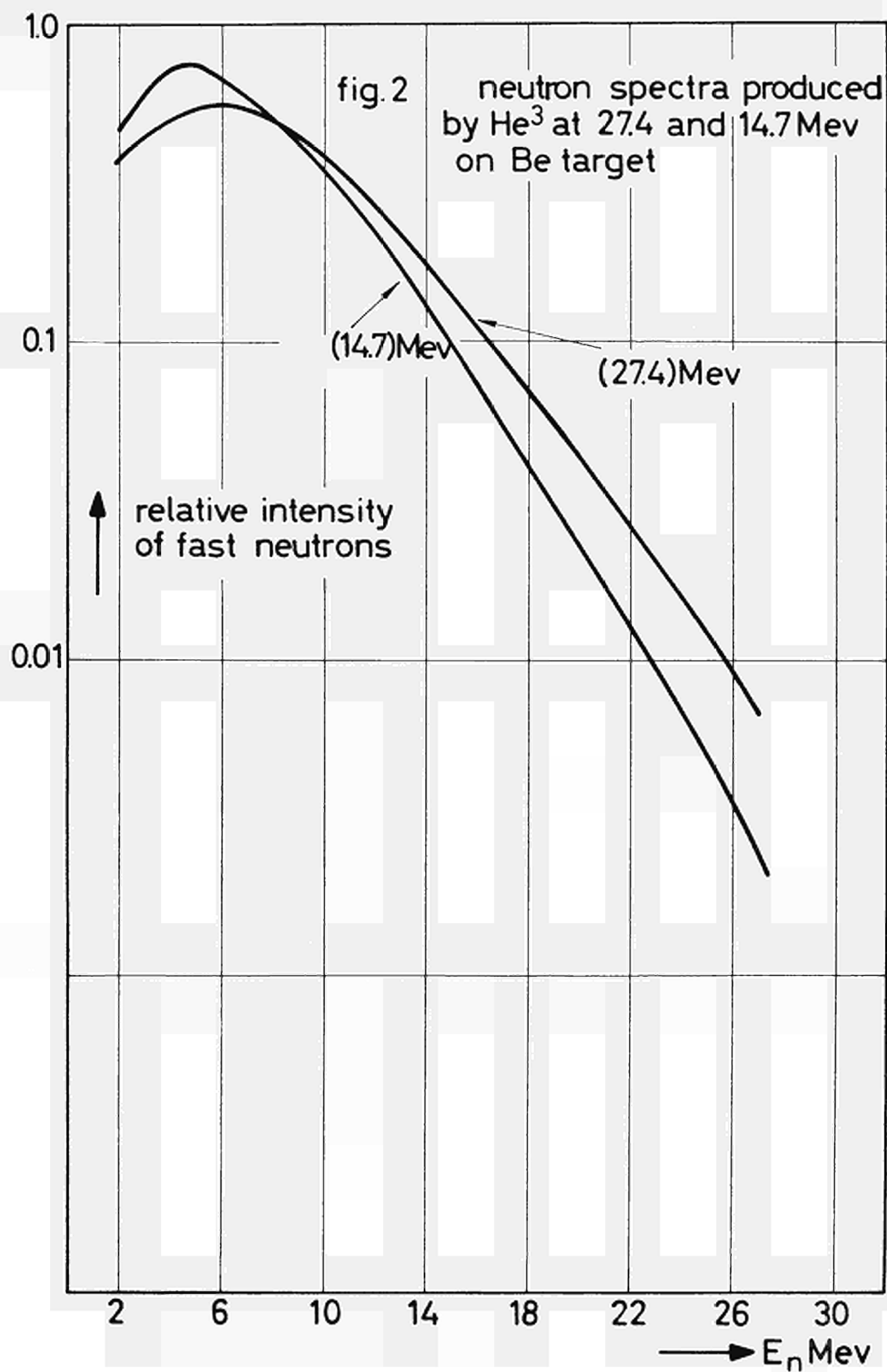


Table III gives a comparison between the values of A_1 A_2 ... etc. (experimentally measured) and A_1 , A_2 etc.... (calculated by means of formula (4))

Table III

Threshold reaction

	$\text{Si}^{28}(\text{n},\text{p})\text{Al}^{28}$	$\text{Al}^{27}(\text{n},\alpha)\text{Na}^{24}$	$\text{Cu}^{63}(\text{n},2\text{n})\text{Cu}^{62}$	$\text{C}^{12}(\text{n},2\text{n})\text{C}^{11}$	$\text{Al}^{27}(\text{n},\text{p})\text{Mg}^{27}$
A(Exp)	8.54×10^4	1.21×10^4	5.08×10^4	6.11×10^2	1.78×10^4
A(calc)	8.58×10^4	1.25×10^4	5.03×10^4	9.8×10^1	1.78×10^4

B) Angular distribution

In order to calculate neutron yields, knowledge of the angular distribution of the fast neutrons is required. Measurements were taken with 20 MeV He^3 at 7 and 10 cm with Aluminium foils of varying size (18, 10 and 5 m/m). No measurable differences were observed for these different sizes and distances.

From fig.(3) one can observe that the distribution of the fast neutrons is strongly peaked in the forward direction. This is obviously an advantage for all possible fast neutron activations. For comparison we have also plotted the corresponding curves for 15 MeV deuterons, also on beryllium.

C) Total production yield

Following the procedure outlined in par II the neutron flux (φ) has been measured at various distances using detector sandwiches of different sizes in order to rule out possible geometry effects. The results for these detectors were integrated over all angles ¹⁴⁾, taking into account the angular distribution.

Table IV summarizes the results obtained.

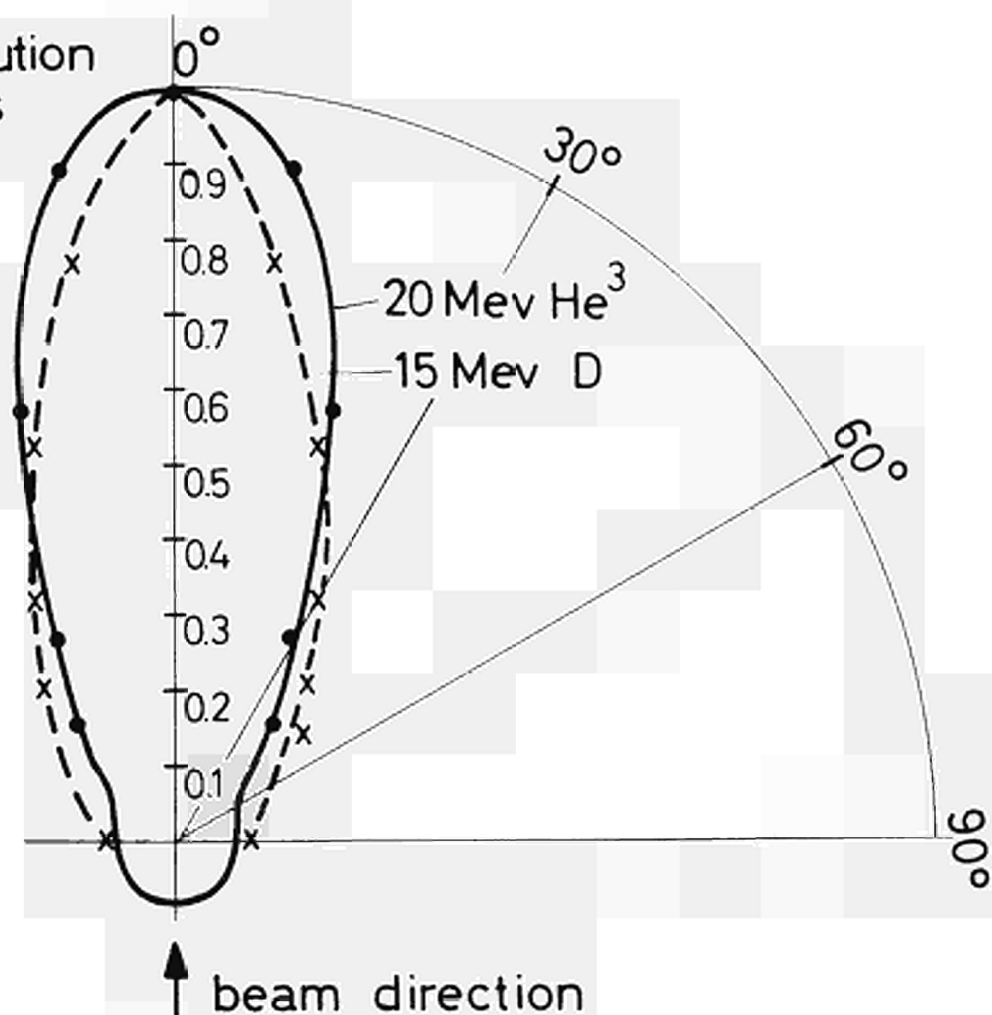
angular distribution
of fast neutrons

fig. 3

TABLE IV

Distance (cm)	Detector diameter (cm)	ϕ (n/sec/ μA)
10	1.8	6.8×10^8
10	1.0	9.6×10^8
7	0.5	14.1×10^8
7	0.5	9.1×10^8
4.5	1.8	5.9×10^8
		AV = 9.1×10^8

Acknowledgements

Numerous discussions with Dr. P. Kramer and the untiring assistance of the AVF cyclotron operators are gratefully acknowledged.

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DISCUSSION

Mr. RICCI

What distance there is between the insulator and the Be target in your Faraday cup ?

Mr. BRUNINX

12 - 15 cm .

Mr. JIGGINS

What is the thickness of the target foils used,
was any correction necessary for self absorption effects ?

Mr. BRUNINX

1) The thickness of the target foils was :

Cu : ~ 1.100 mg / diameter 18 mm

Al : ~ 170 " " " "

Si : ~ 200 " " " "

C : ~ 800 " " " "

2) We did not apply any correction for selfabsorption effects as we measured by γ or coincidence methods.

A correction for possible absorption of the neutrons in the sandwich was also not applied. Indeed in an experiment where copper discs with a thickness of 10 and 20 mm was placed between the Be-target and the sandwich, only a small absorption effect was noticed. As the targets of the sandwich were much thinner, no correction for this effect was applied.

Mr. RICCI

It is not necessary to make correction for flux depression in the foils because these are fast neutrons and cross section for them are of the order of only a few hundreds of millibarns at the most.

Mr. SMITH

Did you measure the neutron spectrum at any angle other than 0° to the

deuteron beam, since, if there is a big variation in the spectrum with angle, a threshold detector would not give an accurate measurement of your neutron yield.

Mr. BRUNINX

We measured a spectrum at an angle of 40° . The spectrum of the fast neutrons did not differ markedly from 0° spectrum (within 10 - 15%). We also measured the angular distribution. With Cu foils (high threshold) no appreciable deviation was found from the angular distribution measured with Al-foils.

Mr. SMITH

At AWRE we use nuclear photoplate emulsions to measure fast neutron spectra. Had you considered using these.

Mr. BRUNINX

We considered the photographic plate method but since the equipment and the technique were quite involved, we did not give it further attention.

Mr. MARKOWITZ

We have measured the $\text{Be}^9 (\text{He}^3, n) \text{C}^{11}$ excitation function by detecting the C^{11} positron radioactivity, and calculate 10^{11} n/sec for a 100 μA beam, in agreement with your measurement of spectra and angular distribution. It is fun to have international agreement.

At 20 MeV, you are on the decrease of the (He^3, n) excitation function and are producing neutrons also by $(\text{He}^3, 2n)$ and perhaps $(\text{He}^3, 3n)$ reactions.

Have you looked at the yields at lower He^3 energy, e.g. 8 or 12 MeV ?

I would guess that while the yield might decrease, such decrease might not be too drastic.

Mr. BRUNINX

We have made an absolute determination at 11,5 MeV and the value is somewhat lower, I believe it is a factor of 2 or so.

Mr. RICCI

We also agree with Dr. MARKOWITZ'S and Mr. BRUNINX'S neutron-output data. Thick targets lead to higher neutron outputs if higher energies are used. This is because at higher energies there is reaction with low as well as

high energy particles. In other words all energies are represented; the range is larger.

Mr. SCOTT

I am a little surprised that Mr. BRUNINX should prefer to use a foil activation method rather than a manganese bath technique. It would seem that there are a large number of intermediate steps and assumptions which have to be made between the activation and the calculation of the yield per μa which can lead to errors, and I think that the results shown support this fact. At Birmingham University we are interested in measuring the neutron yield from a thick lithium target (using 10 MeV protons) and are using a continuous flow manganese bath. The normal trouble with measuring accelerator yield is, that the beam current is not stable over long periods of time but if one examines the equations governing the growth of activity in the continuous flow technique it is possible to write them such a way that any variations in beam current can easily be taken into account.

Mr. BRUNINX

I agree with Dr. SCOTT that the manganese bath technique is able to give more accurate, precise answers, but as we wanted to know both the spectrum and the total production rate, we did not use the manganese bath, as it gives only a value for the flux, not the spectrum. We are comparing however values obtained by the manganese bath, threshold detectors, and calibrated neutron sources for 18 MeV protons on Be.

THE FAST NEUTRON FACILITY AT THE PRETORIA CYCLOTRON

W.J. Naudé^{*}, Max Peisach^{**} and W.L. Rautenbach

Nuclear Physics and Radioactivity Division, National Research Laboratory
Pretoria, South Africa

^{*} The Merensky Institute for Physics, University of Stellenbosch

^{**} Southern Universities Nuclear Institute, Faure C.P.

ABSTRACT

The construction of a beryllium target for use with the internal beam from the Pretoria 110-cm. cyclotron, as a neutron source, is described. The beryllium is mounted on a copper support through which a rapid current of cooling water flows. The transfer of heat is improved by grooving the underside of the target and by spreading the charged particle beam over the entire surface.

Samples for neutron irradiation are mounted in a cylindrical pipe in six positions from 6.5 to 23.6 cm. from the beryllium and at angles from 14° to 70° to the direction of incidence of the charged particle beam. The pipe protrudes through the vacuum lock so that samples can be irradiated outside the vacuum system of the cyclotron.

Activation reactions with thresholds between 1.8 and 20.3 MeV. were used to calibrate the neutron flux. Ten reactions were so chosen that it was possible to use only 5 target materials, viz., KOH, Al, MgO, NH_4F and C. Thus only 6 irradiations were sufficient to measure the flux at each irradiation site. The activities were measured with a NaI(Tl) scintillation detector, the efficiency of which had been calibrated as a function of gamma-ray energy.

Flux calibrations were performed on the neutrons emitted in the reaction ${}^9\text{Be}(d,n){}^{10}\text{B}$ with 15.7 MeV. deuterons and the reaction ${}^9\text{Be}(\alpha,n){}^{12}\text{C}$ with 31.8 MeV. alpha particles.

Total fluxes, depending on the irradiation site, ranged from 1.8×10^8 to 2.1×10^{10} neutrons/cm.²-sec. The energy distribution was measured and found to be in excellent agreement with published data obtained from 15, 24 and 26 MeV. deuterons.

Agreement was also good when the angular distribution of the emitted neutrons was compared with previously reported values. Total fast neutron emission from deuteron bombardment at 15.7 MeV. was 2.28×10^{12} neutrons/sec. per 100 μ A., which may be compared with the reported value of 1.90×10^{12} neutrons/sec. at 15 MeV.

The Pretoria 110-cm. cyclotron (1) can produce internal and external proton, deuteron and alpha-particle beams with maximum energies of about 8, 16 or 32 MeV. respectively, and is capable of producing internal beam currents of up to 800 μ A of hydrogen ions or 200 μ A helium ions. External beams with well-defined energy (within $\pm 0.3\%$) can be obtained at currents of a few microamperes. The large difference between internal and external beam currents makes it attractive to use the internal beam for neutron production.

Measurements by other authors (2) have shown that the total neutron yield obtainable from the irradiation of thick targets with accelerated deuterons or alpha-particles increases rapidly with decreasing atomic number of the target. Moreover, the angular distribution of the neutron flux from light elements has a pronounced peak in the forward direction. It follows that the optimum ratio of useful neutron flux to charged particle beam current is obtainable in a position as close as possible to a light element target, in the direction of the incoming charged particle beam.

Because the neutron flux obtainable from a target is directly proportional to the incident charged particle beam current, the practical limitation to the intensity of the neutron flux is imposed by the thermal properties of the target which determine the rate of removal of heat from the region irradiated. The light element, beryllium, has thermal properties which meet

the requirements for neutron production; it has very good thermal conductivity, a comparatively high melting point and low vapour pressure over a wide temperature range, thus making it useful for a target material inside the vacuum of the cyclotron.

The paper describes the beryllium target used at the Pretoria cyclotron and the calibration of the flux and energy distribution of neutrons obtained from it by irradiation with 15.7 MeV. deuterons and 31.8 MeV. alpha-particles.

DESCRIPTION OF THE TARGET AND IRRADIATION ASSEMBLY

The target consisted of a block of beryllium soldered onto a copper plate (see Figure 1), by placing a foil of silver, 0.1 mm. thick, between the copper and beryllium, and heating the system, under an inert atmosphere, to 900°C. The copper plate was, in turn, silver-soldered onto a copper block through which the cooling water circulated rapidly. To improve heat transfer from the copper plate to the water, the area of contact was increased by grooving the underside of the copper plate. The entire target head could be fitted onto a standard target support, thereby making it a simple operation to change target dimensions and materials.

By selecting the correct angle between the beryllium surface and the target support, a well-centred cyclotron beam could be spread over the entire surface of the beryllium target

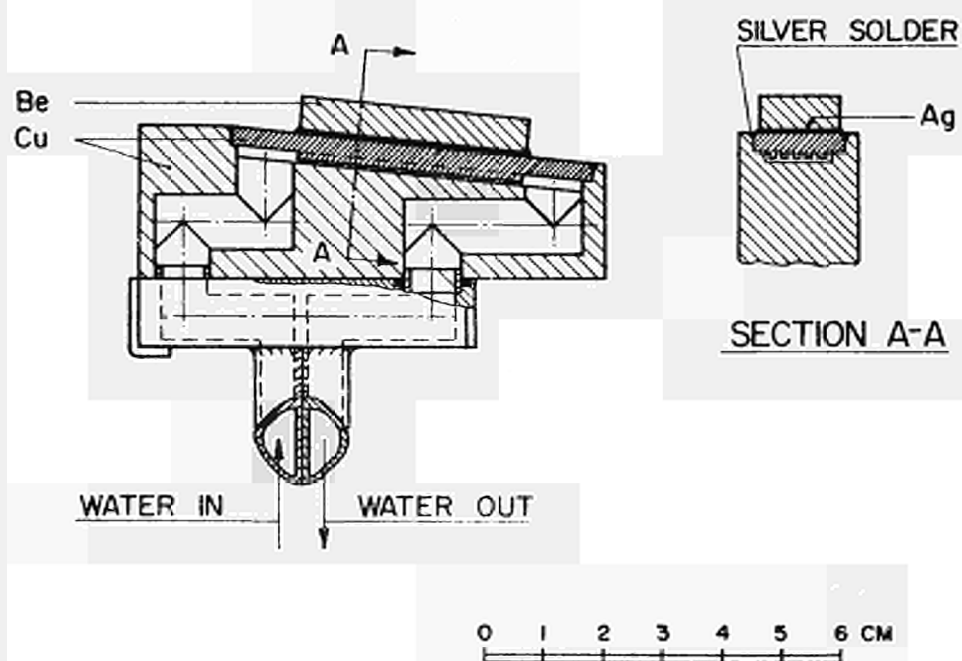


Figure 1. The construction of the beryllium target head.

whereby the heat generated could be dissipated over a larger area. The centering of the beam was achieved by adjusting the current through the first harmonic coils of the cyclotron so as to obtain equal beam currents onto the beryllium target and onto an auxiliary target placed at the same radius from the centre of the cyclotron, 180° away (see Figure 2). When the auxiliary target was retracted a short distance, the entire internal beam fell on the beryllium. Erroneous measurements of the beam current due to secondary electron emission was prevented by copper shielding plates above and below the targets.

Samples for neutron irradiation were positioned in a cylindrical brass pipe, closed at one end and situated immediately behind the beryllium target (see Figure 3). The open end of the pipe protruded through the vacuum lock through which the entire assembly could be introduced into, and removed from, the cyclotron. The sample holder, into which the samples were mounted, fitted into the pipe and could be inserted into any depth required, and withdrawn rapidly. The advantage of such an assembly lies in the fact that the sample is irradiated at atmospheric pressure, thus simplifying sample preparation and irradiation procedure. A fast pneumatic transfer system can readily be incorporated. Since the brass pipe is situated within the radio frequency field of the cyclotron, and because the danger exists that a small fraction of the circulating cyclotron beam may strike the tube, a jet of compressed air was

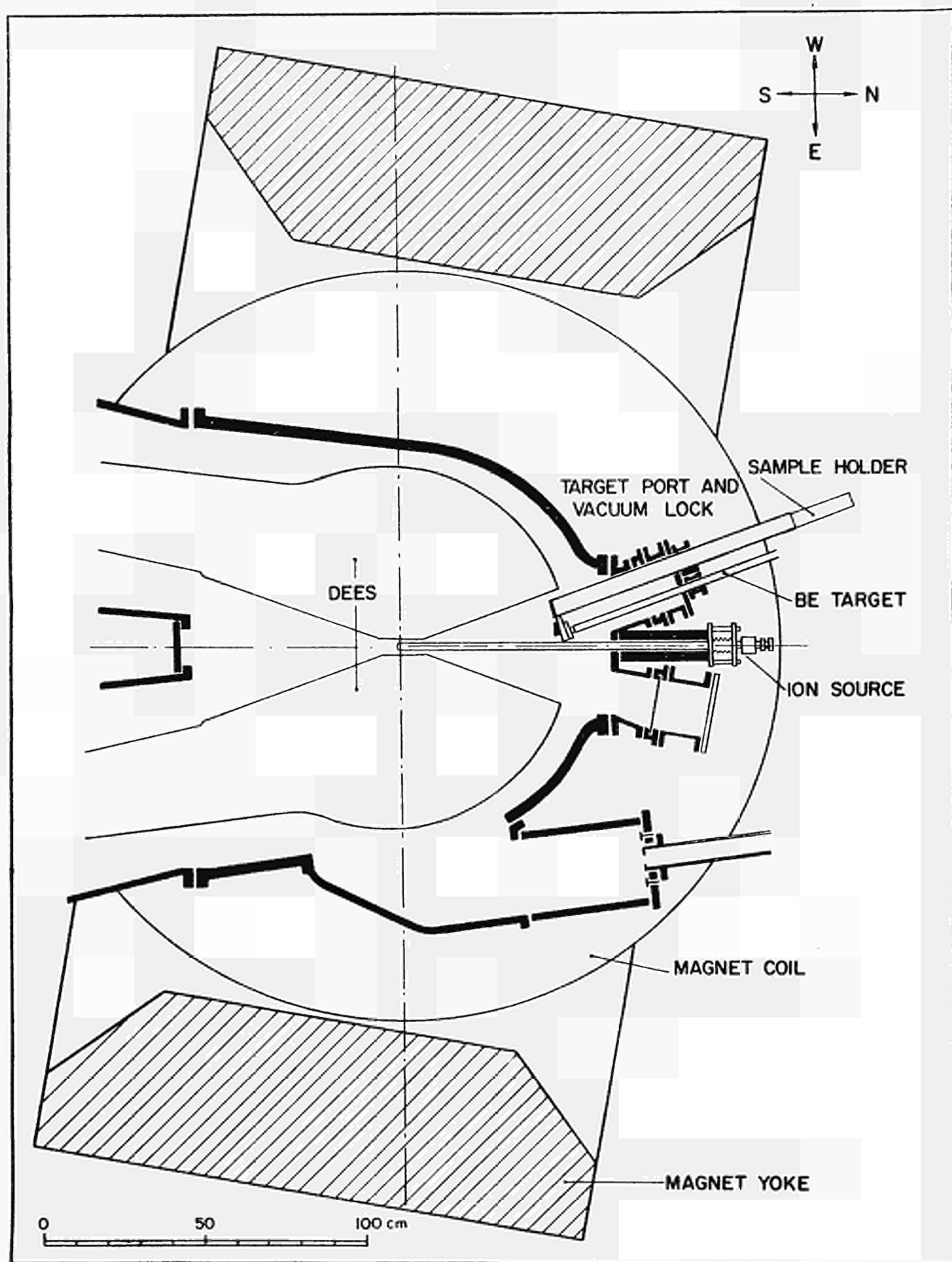


Figure 2. The position of the neutron facility in the cyclotron.

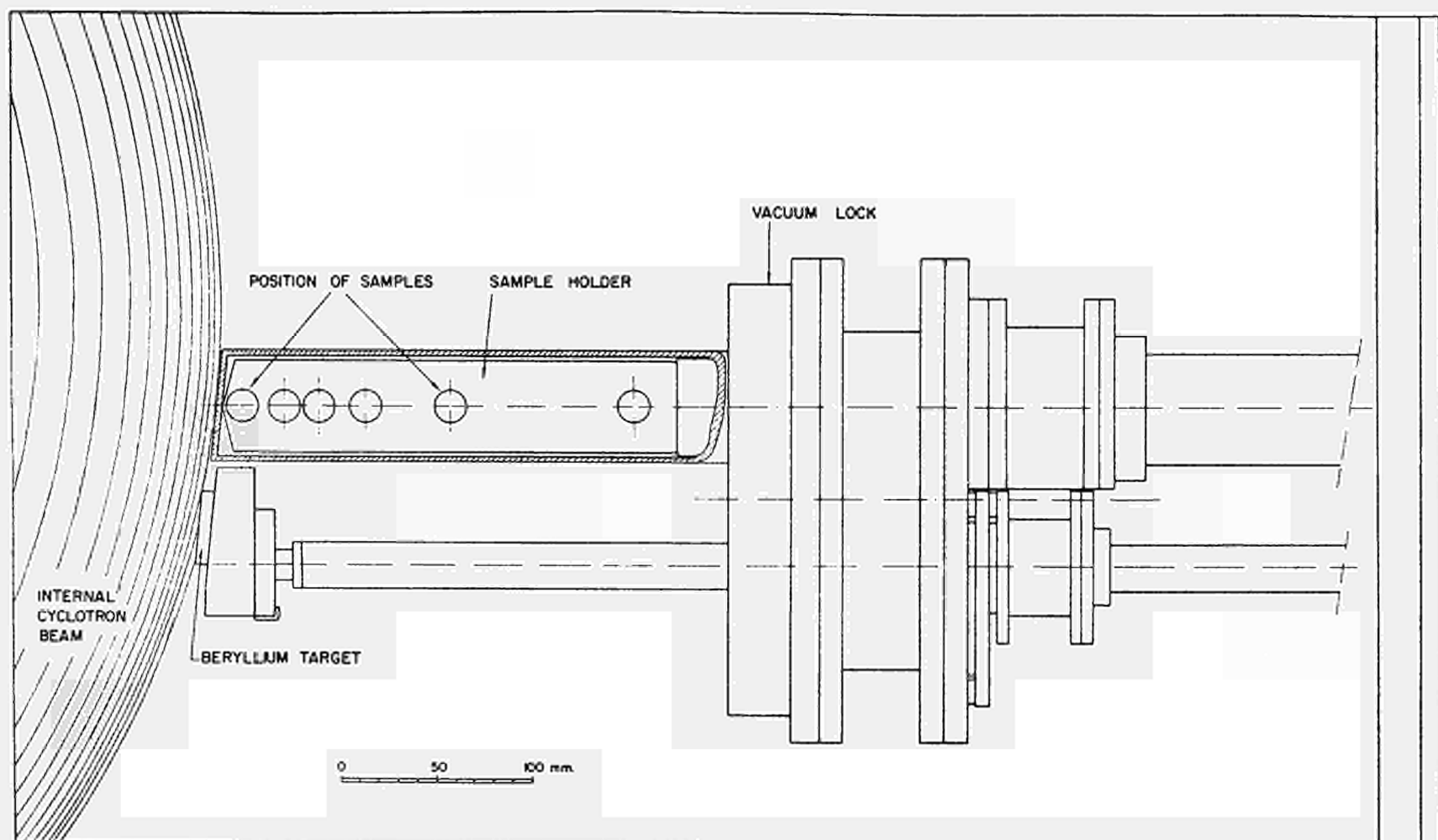


Figure 3. The neutron irradiation assembly.

usually directed onto the closed end in order to cool the pipe and to provide air cooling for the irradiated samples, if necessary.

CALIBRATION OF NEUTRON FLUX

The use of activation threshold detectors still represents a useful technique for measuring neutron fluxes and their energy distribution. This method was used to determine the flux and energy distribution of neutrons produced by 15.7 MeV. deuterons and 31.8 MeV. alpha particles incident on a thick beryllium target.

Selection of reactions with suitable thresholds

Neutrons with energies up to about 20 MeV. were expected from bombarding the target with deuterons and up to about 35 MeV. from the alpha particle irradiation. It was thus necessary that the reactions selected for calibrating the neutron flux should have thresholds ranging over as wide an energy as possible within the expected neutron energy range. Furthermore, reactions had to be selected for which the excitation curves were available, at least in part, and from which the activities that were produced were readily observable. The half-life of the products could not have been too short, as this would have led to difficulties in transporting the activated sample to the counting assembly, nor too long, requiring a

comparitively long irradiation time. In addition it was desirable that some target nuclides should be chemically combined in stable and readily available pure compounds so that results of more than one reaction could be observed from a single irradiated sample.

The reactions that were selected are given in Table I together with the half-life of the product concerned in each case and the energies of important gamma-rays that were used to measure the activities. The 10 listed reactions were selected because they met the requirements outlined above. All the reaction products could be observed by the irradiation of only 5 target materials, KOH, Al, MgO, NH_4F and C, which were counted with a single counting assembly, at appropriate times after the end of the activation, even when all five target materials were simultaneously activated. Reference to Figure 3 shows that the six irradiation positions could thus be simultaneously calibrated, in a series of only six irradiations during which every target material was irradiated in every position and a blank container could be accommodated in every position in turn to allow for subtraction of unwanted activities generated in it.

The only drawback with the abovementioned set of reactions lies in the fact that no reaction is included with its threshold between 5 and 11 MeV. However, the advantages of the selected reactions in other ways were so great as to outweigh this disadvantage.

TABLE I
NEUTRON THRESHOLD REACTIONS

Reaction	Approximate threshold (MeV)	Half-life of product	Energies of important gamma-rays* (MeV)
$^{41}\text{K}(n,p)^{41}\text{Ar}$	1.8	1.83 h.	1.29
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	1.9	9.5 m.	0.84, 1.01
$^{41}\text{K}(n,\alpha)^{38}\text{Cl}$	3.3	37.3 m.	1.62, 2.16
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	4.0	15 h.	1.37, 2.75
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	4.9	15 h.	1.37, 2.75
$^{19}\text{F}(n,2n)^{18}\text{F}$	11.0	110 m.	(β^+)
$^{14}\text{N}(n,2n)^{13}\text{N}$	11.4	10 m.	(β^+)
$^{39}\text{K}(n,2n)^{38}\text{K}$	13.4	7.7 m.	(β^+), 2.16
$^{16}\text{O}(n,2n)^{15}\text{O}$	16.6	124 s.	(β^+)
$^{12}\text{C}(n,2n)^{11}\text{C}$	20.3	20.5 m.	(β^+)

* (β^+) refers to positron emission and hence the appearance of 0.51 MeV. gamma-rays.

Irradiation and measurement

Weighed quantities of the selected target materials were irradiated in polyethylene vials of 3 cm.³ capacity. The polyethylene containers were mounted in each of the six positions shown in Figure 3. These positions correspond respectively to directions of 14, 31, 40.5, 50, 60 and 70° relative to the incident charged particle beam, calculated with respect to the centre of the beryllium target and the midpoint of the irradiated samples, and were at distances of 6.5, 7.5, 8.7, 10.6, 14.5 and 23.6 cm. respectively.

The beryllium target was irradiated with internal beam currents of about 100 μ A. for 10 minutes and the total current falling on it was measured with a current integrator. Additional monitoring was carried out using cylinders of copper, placed about a meter from the beryllium target, in which the relative yields of both copper-62 and copper-64 from the reactions $^{63}\text{Cu}(n,2n)^{62}\text{Cu}$ and $^{65}\text{Cu}(n,2n)^{64}\text{Cu}$ respectively, were compared with the current integrator values. For intercomparison between different irradiations, all values were normalised to a total charge of 60 millicoulombs, but the normalisation factors were always within $\pm 10\%$ of unity. The neutron-induced activities in the samples and the copper monitors were measured with a 3 x 3" NaI(Tl) scintillator crystal the efficiency of which was calibrated using substandards of ^{137}Cs , ^{84}Mn , ^{60}Co and ^{22}Na . Counting of the samples was in most cases continued for several half-lives of the product nuclide, from which, the

activities at the end of the irradiation were deduced.

Flux calibrations

The excitation curves for the reactions used for calibrating the neutron flux are shown in Figures 4 and 5, where the solid curves are based on published data (3,4) and the dotted lines are the assumed shapes in the region where experimental data were not available. The integral neutron flux, as obtained from the activation reactions on samples placed in positions 3 and 6 are given as examples, in Figure 6, and in position 1 in Figure 7, where deuterons and alpha particles were respectively used to generate the neutron flux. These figures both show the variation of the neutron flux, for neutrons of energy greater than \underline{E} , with neutron energy, \underline{E} .

The energy distribution of the neutrons from the reaction ${}^9\text{Be}(\text{d},\text{n}){}^{10}\text{B}$ is shown in Figure 8 for all the irradiation positions, and for the reaction ${}^9\text{Be}(\alpha,\text{n}){}^{12}\text{C}$ in Figure 9 for irradiation position 1.

The results shown in Figures 6-9 were calculated by an iteration process for which initially it was assumed that the energy distribution of the neutrons was independent of neutron energy. Three iterations were sufficient to reproduce the reported values.

The shape of the energy distribution curves in Figure 8 is in excellent agreement with previous work (5,6,7) reported for incident deuteron beams of 15, 24 and 26 MeV., especially

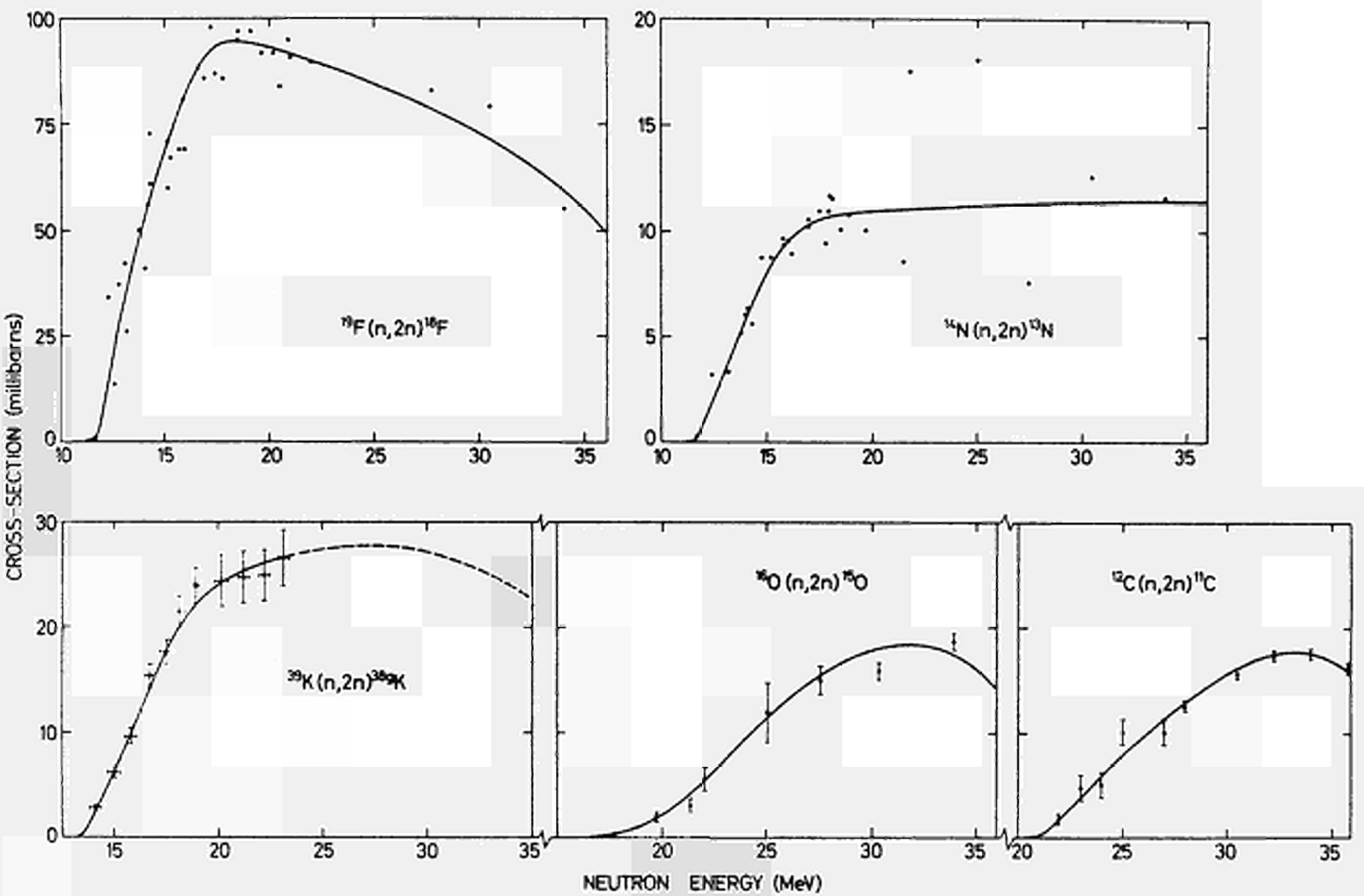


Figure 4. Excitation curves for some (n,2n) reactions used to determine the neutron flux. The broken line shows the assumed shape of the curve in a region of insufficient data.

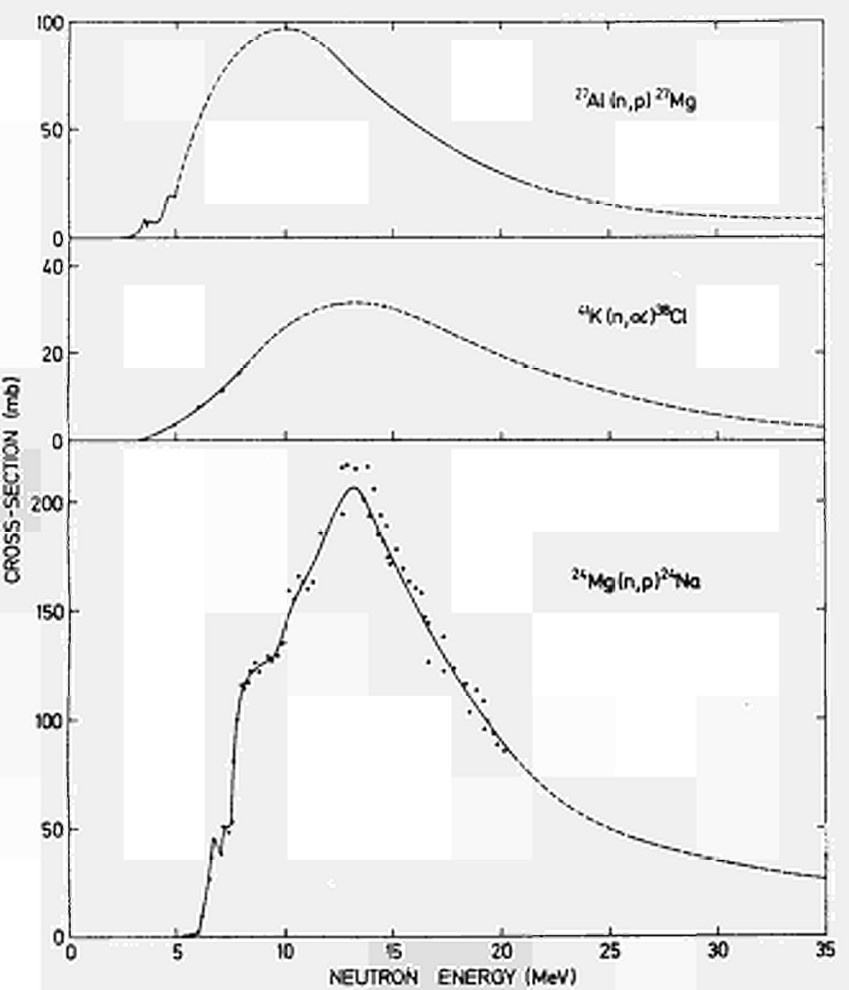
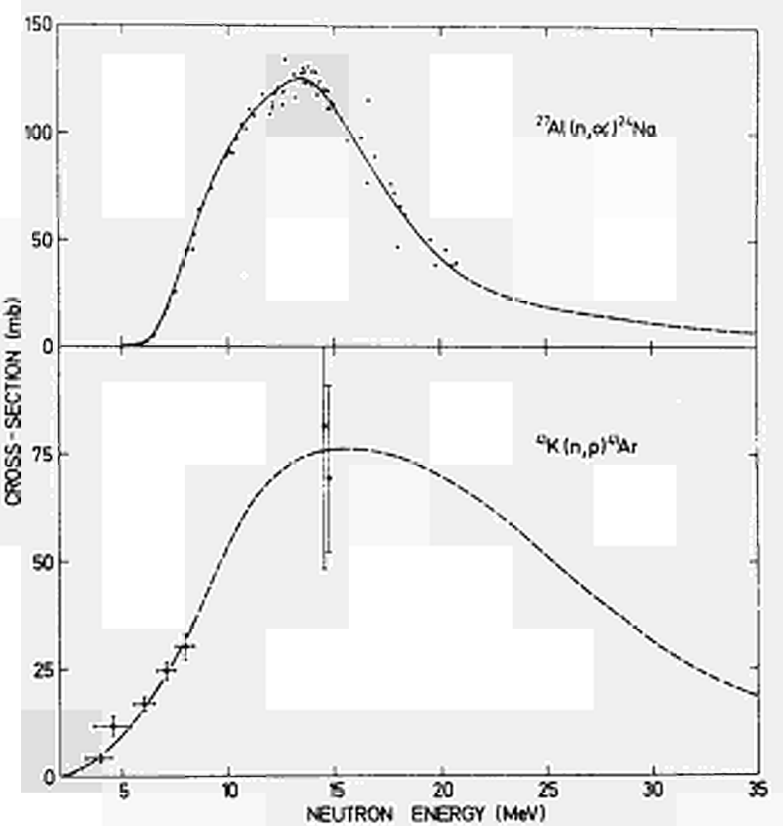


Figure 5. Excitation curves for some (n,p) and (n, α) reactions used to determine the neutron flux. The broken lines show assumed shapes of the curves in regions of insufficient data.

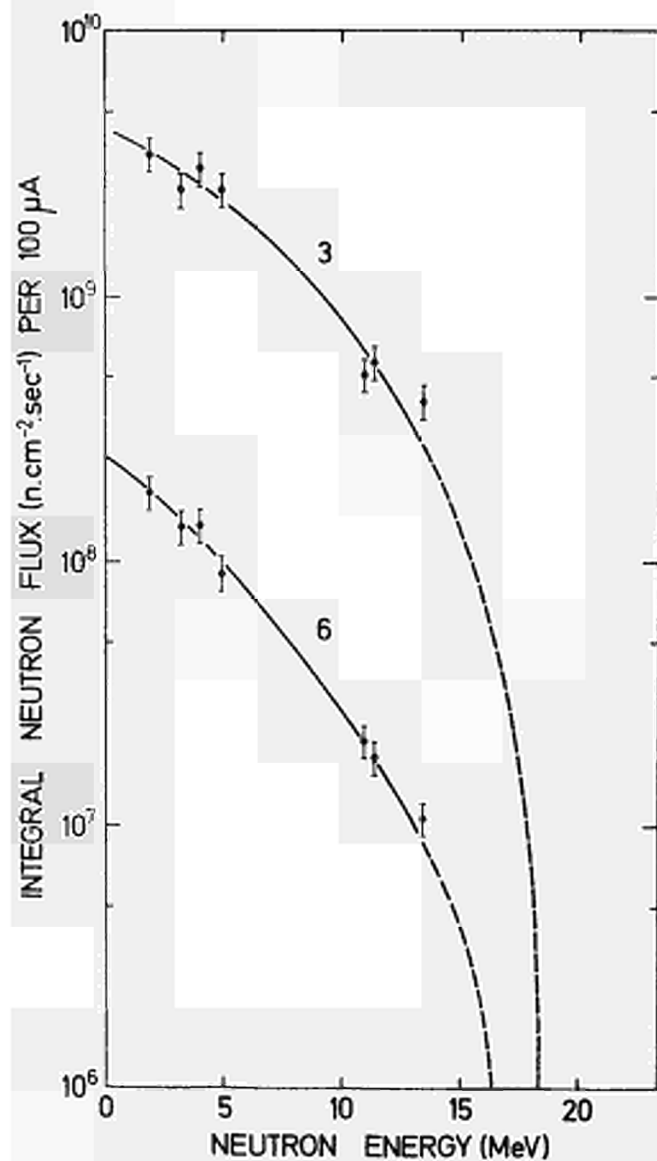


Figure 6. The variation with the neutron energy E , of the total neutron flux, for neutrons with energy greater than E , as obtained at irradiation positions 3 and 6 from the reaction ${}^9\text{Be}(d,n){}^{10}\text{B}$.

$$E_d = 15.7 \text{ MeV.}$$

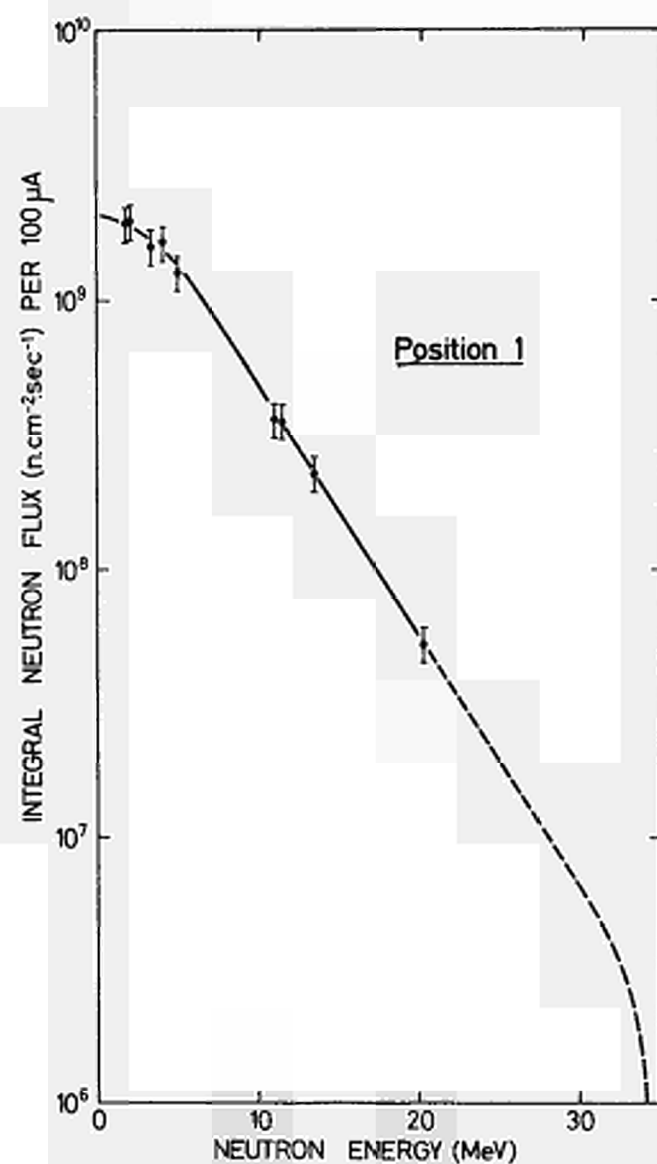


Figure 7. The variation with the neutron energy E , of the total neutron flux, for neutrons with energy greater than E , as obtained at irradiation position 1 from the reaction ${}^9\text{Be}(\alpha,n){}^{12}\text{C}$

$$E_\alpha = 31.8 \text{ MeV.}$$

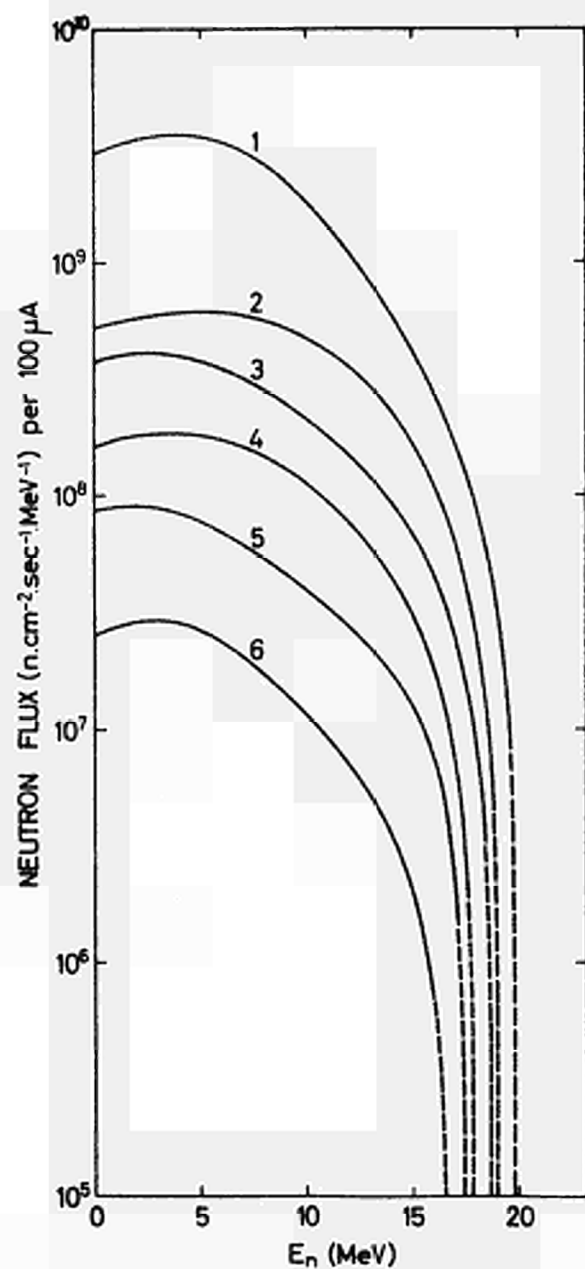


Figure 8. Energy distribution of neutrons from the reaction ${}^9\text{Be}(d,n){}^{10}\text{B}$ at all six irradiation positions.

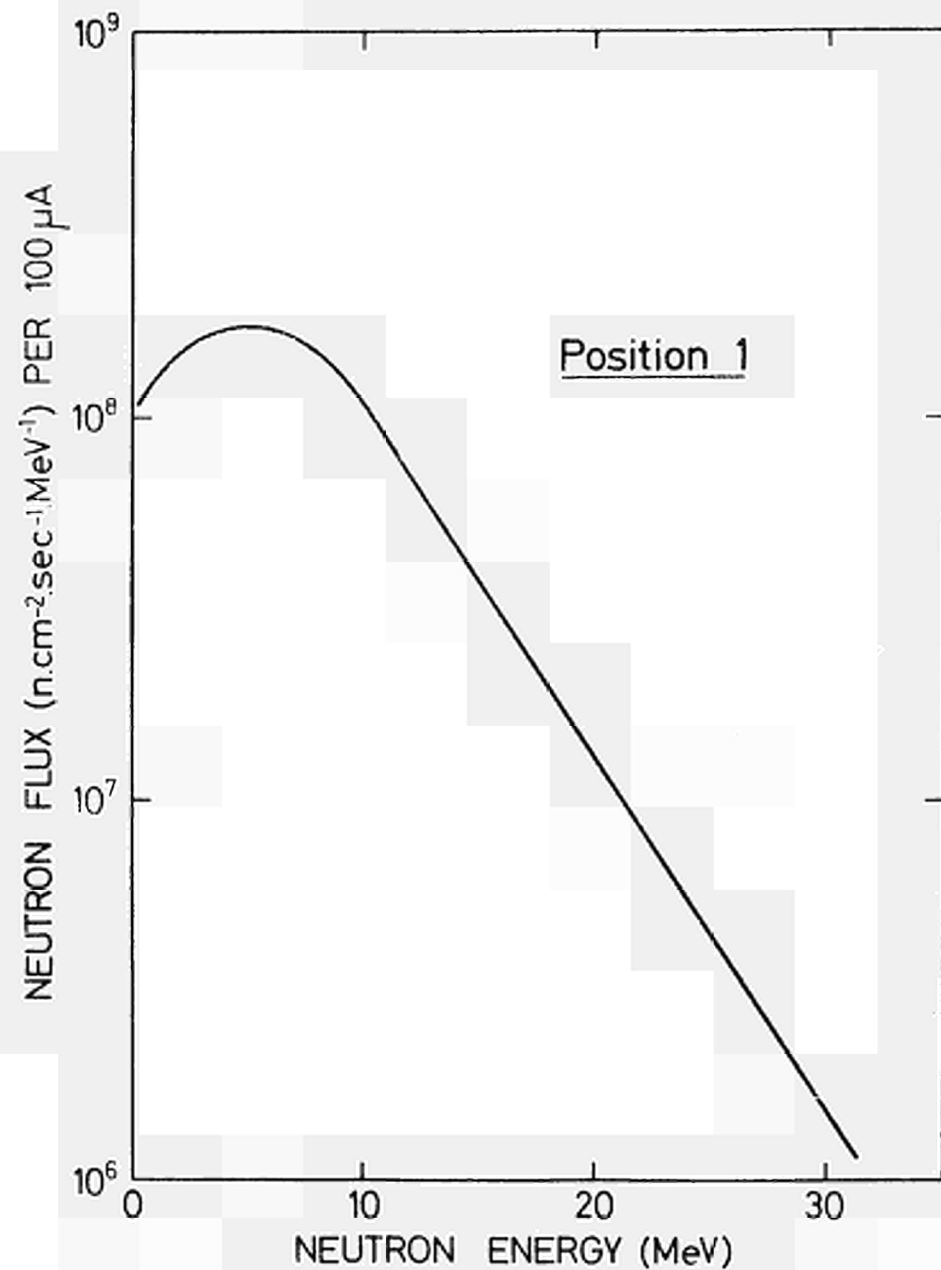


Figure 9. Energy distribution of neutrons from the reaction ${}^9\text{Be}(\alpha,n){}^{12}\text{C}$ at irradiation position 1.

when allowance is made for the difference in bombarding conditions. In the case of the reported data for 15 MeV. deuterons (5), the rate of flux decrease with neutron energy was somewhat greater than that observed in this work, but as the conditions of the deuteron bombardment were not defined, quantitative comparison between the reported (5) and observed results was not possible.

The angular distribution of the neutron flux can be deduced if correction is made for the different distances between the irradiation positions and the centre of the beryllium target. However, in this connection the errors in determining the mean angle between the target and the internal cyclotron beam incident on the beryllium, arising from the fact that neither the beryllium target, nor the irradiated samples, can be considered as points, become large, especially with decreasing angle. Furthermore, the uncertainty in the actual distance between the sample and some mean point of neutron generation in the beryllium introduces very large errors in the relative neutron flux intensity at small angles. The sizes of the probable errors from these sources are marked in Figure 10 where the experimental results are shown as points and are compared with the reported (2) angular distribution of neutrons

from the reaction ${}^9\text{Be}(\text{d},\text{n}){}^{10}\text{B}$, shown as a smooth curve.

When the observed angular distribution from 0° to 90° (see Figure 10) and the reported data (2) from 90° to 180° were used to calculate the total neutron emission with energy above 1 MeV. from a thick target of beryllium by a deuteron beam of 15.7 MeV., the value obtained was 2.28×10^{12} neutrons/sec. per 100 μA . This value should be compared with the corresponding result (2) for 15 MeV. deuterons of 1.90×10^{12} neutrons/sec. per 100 μA .

ACKNOWLEDGEMENTS

We would like to thank the Director of the National Physical Research Laboratory for the use of the cyclotron and the Director of the National Chemical Research Laboratory for supplying funds to cover travelling expenses of one of us (M.P.). The use of some equipment from the Southern Universities Nuclear Institute is gratefully acknowledged. Carl Verwey, Kobus van der Merwe and the operators of the Pretoria cyclotron are thanked for their valued assistance during the experiments. Hugh Schmitt and E. Thain prepared the drawings in this manuscript.

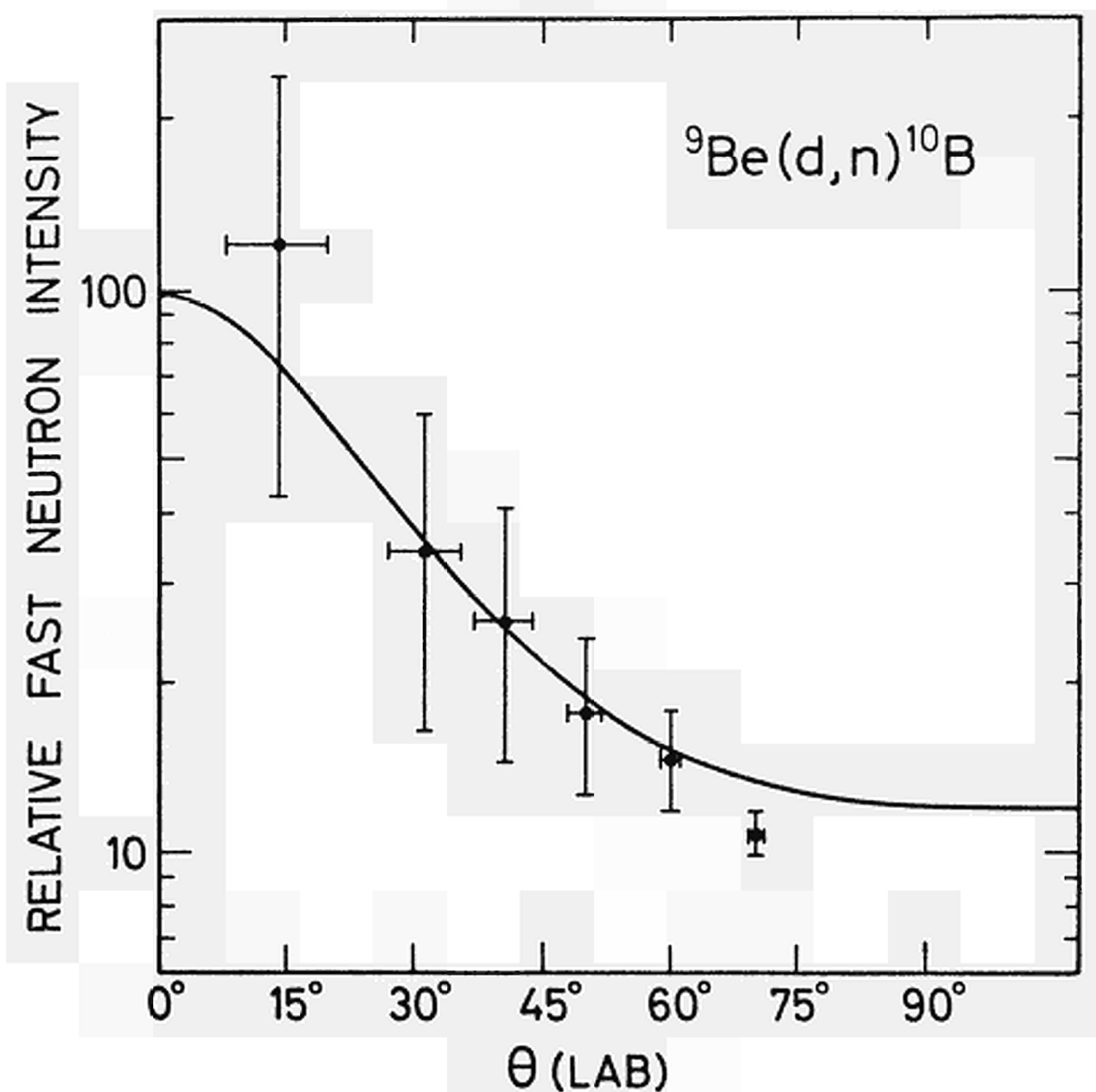


Figure 10. Angular distribution of the fast neutron flux.

The points are experimentally determined and the curve represents reported results (2).

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DISCUSSION

Mr. MARKOWITZ

It may not be quite fair to ask about the choice of experimental method after your fine work with threshold detectors, but I do wonder about the rejection of proton-recoil scintillation spectrometry in your work or that of the previous speaker, Dr. BRUNINX. Is gamma-ray interference the reason ?

Mr. PEISACH

The space in the irradiation pipe holding the samples was so limited that proton-recoil detectors were difficult to mount. We also have little experience with that kind of measurement.

Mr. KUIN

Why do you use the internal and not the external beam ?

Mr. PEISACH

The external beam has already been adapted to high resolution physics experiments so that the total current obtainable externally was much smaller than the internal one. We thus had very little choice.

The use of the Manganese Bath Technique for
measuring the neutron yield of accelerator sources

M. C. Scott

Department of Physics,
University of Birmingham.

ABSTRACT

The use of the continuous flow manganese bath technique to measure the yield of sources which vary with time in some known manner is described, and the results of tests on the method are given.

The Nuffield Cyclotron at Birmingham University is being used increasingly for neutron production in a range of experiments. We are particularly interested in using it for obtaining cross section data and for doing integral spectrum measurements, and for both types of experiment we need to know the yield and spectra of the targets used as accurately as possible (we are at present using thick lithium targets with 10 Mev protons and beam currents of up to 20 μ A). In this paper I should like, very briefly, to outline the method

being used to measure the yield, using the manganese bath technique.

You will all, I am sure, be familiar with the principles of this method, so I will not go into any details here. You may not, however, be quite so familiar with the continuous flow method described by AXTON, CROSS and ROBERTSON of the National Physical Laboratory, Teddington, in which the active solution of manganese sulphate is pumped continuously from the sphere containing the source to a counting chamber which may be situated as far away as is convenient. This means that the detector, in our case a sodium iodide scintillation counter, can easily be shielded from gamma rays produced by the source. The count rates during both the growth and decay of activity are used to predict the asymptotic (saturation) count rate.

The original analysis of Axton et al was only for a static source, but many accelerators, and in particular cyclotrons, do not have a beam current which is steady over periods of the order of hours, so that it is necessary to make an allowance for effect of these fluctuations in source strength on the measured activity if the method is to be used on accelerator sources. If $A(t)$ is the number of active atoms of

Mn^{56} , in the sphere at a time t after the start of the activation the equation governing the growth of activity is, assuming that the mixing is instantaneous,

$$\frac{dA(t)}{dt} + A(t) \left[\lambda + \frac{C}{pV} \right] - \frac{C}{pV} e^{-\lambda p} A(t-p) = FQ(t) \quad (1)$$

where

- λ = decay constant of Mn^{56}
- C = volume of counting system and associated pipe work
- V = volume of the sphere containing the source
- p = pumping time, the time taken for the solution to be pumped to the counting chamber and returned to the sphere
- $Q(t)$ = source strength at time t
- $F = f(1-L)(1-m)$, the fraction of neutrons captured in Mn^{55} in which
 - L = neutron leakage fraction
 - m = fraction of neutrons captured in the source and mounting
 - f = fraction of neutrons not lost which are captured by the Mn^{56}

The second term in this equation includes the loss of active atoms pumped out to the counting chamber, while the

third term represents the gain of active atoms pumped back; it is this term which complicates the solution of the equation.

However, we can write the equation in a more convenient form if we consider the solution in a time interval $(n-1)p \leq t < np$, which we shall call the n^{th} interval. If we call this solution $A_n(t')$ then the equation becomes

$$\frac{dA_n(t')}{dt'} + uA_n(t') = FQ(t') + \gamma A_{n-1}(t'-p) \quad (2)$$

where γ and u are the appropriate constant terms in equation (1). Integrating equation (2) from $t' = (n-1)p$ to t we get

$$A_n(t) = e^{-ut} \left\{ A_n[(n-1)p] e^{pu(n-1)} + \int_{(n-1)p}^t e^{ut'} \gamma A_{n-1}(t'-p) dt' + \int_{(n-1)p}^t e^{ut'} FQ(t') dt' \right\} \quad (3)$$

We now note that, if the activation is started at $t = 0$, $A(t) = 0$ for $t \leq 0$. Thus, in the first interval the term $A(t-p)$ vanishes, and the solution for $A_1(t)$ can be written down immediately when the appropriate source strength variation is put into the integral. While the solution in successive intervals can now be continued in a stepwise manner, since

the solution in the preceding interval will be known, the analytical form of the equation clearly becomes very complex after only a few intervals. However, the form of equation (3) is such that the equation can readily be evaluated numerically, the numerical solution for periods of several hours taking only a few minutes of computer time.

If $C(t)$ is the count rate measured in the detector at a time t , and the latter is assumed to be midway between the exit and inlet to the sphere, then the predicted asymptotic count rate C_{∞} is given by

$$\frac{C_{\infty}}{A(\infty)} = \frac{\bar{C}}{\frac{1}{\beta} \int_T^{T+\beta} A(t-p/2) dt} \quad (4)$$

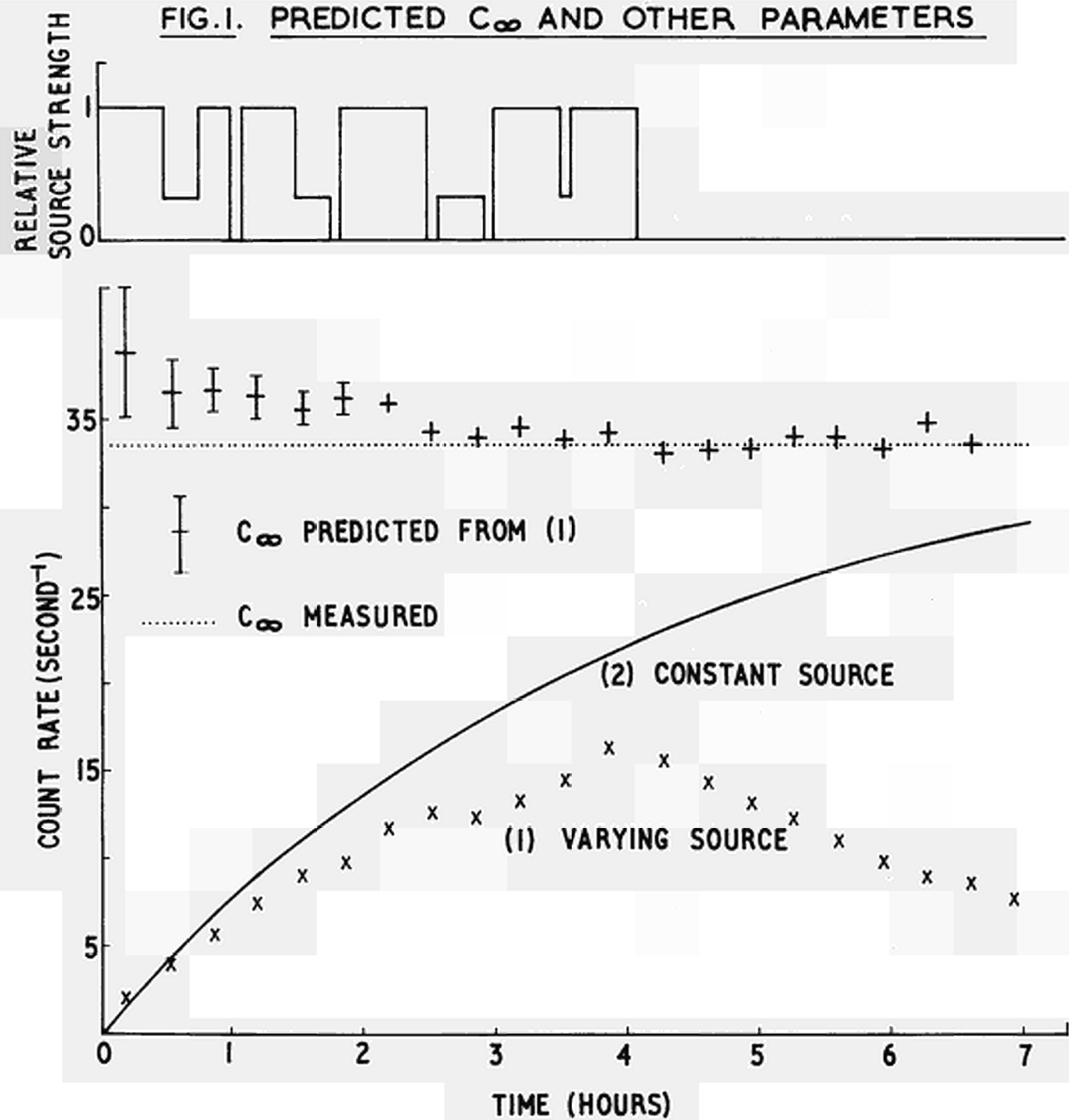
where \bar{C} is the mean count rate in the counting interval T to $(T+\beta)$. If the source strength is normalised to that at the start of activation, $A(t)$ can readily be found from the solution of equation (1), and C_{∞} is then the asymptotic count rate which would have been obtained had the source strength remained constant at its initial value.

To test the validity of this method an extreme form of source variation has been simulated using two static

sources whose relative strengths had previously been accurately determined (1 : 0.256), and the results of one of the preliminary tests is shown in figure 1. The way in which the source strength was varied is shown at the top of the figure. At the bottom are shown the count rates for two cases, (1) being for the source variation shown and (2) being for the case where the source strength was not varied, but was of unit strength throughout the run. Finally, in the middle, are shown the values of the asymptotic count rate for the unit source predicted from the varying source results with the value of the measured asymptotic count rate for the unit source shown for comparison.

It can be seen that the predicted asymptotic count rates are, in general, in excellent agreement with the measured one. The slightly higher values for C_{∞} predicted at the start of the run are almost certainly attributable to poor mixing, the effect of which is most evident when the rate of growth of activity is greatest. Although the pumping speeds used are high (~ 1.3 litres/second) and there are three different outlet positions to try and ensure adequate sampling, the system is a very large one (total capacity ~ 360 litres) and it is obvious that more vigorous stirring is necessary. To

FIG.1. PREDICTED C_{∞} AND OTHER PARAMETERS



this end a much larger pump is being installed, with a pumping rate of about 4 litres/second.

However, the fact that the agreement with the measured asymptotic count rate improves with time is evidence that the growth and decay of the manganese activity has been accurately predicted, since any errors in the solution would lead to results which diverged increasingly with time.

When this method is used to determine accelerator source strengths the information on the relative source strength as a function of time can be obtained from, for example, the beam current variation or, as in our case, from a thin scattering foil together with an associated semiconductor detector placed before the target. The latter also enables the beam energy to be determined, since this is varied by placing absorber foils before the main analyser magnet.

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OPTIMISATION OF MODERATORS FOR PULSED NEUTRON TARGETS

B.H. Patrick, E.M. Bowey, M.C. Moxon and E.R. Rae

Atomic Energy Research Establishment
Harwell, Didcot, Berks., England

ABSTRACT

In time-of-flight experiments, the source of neutrons in the energy range below ~ 100 keV is often a moderator placed next to a heavy element target which is injected with short bursts of charged particles. The effect of the moderator is usually to broaden the neutron pulse. Michaudon, in a series of Monte Carlo calculations, has estimated the magnitude of this effect. He has also shown that the moderator can be characterized at an energy E by a "figure of merit" F given by $F = \frac{N}{\sigma^2}$, where N is the number of neutrons leaving the moderator with energy E and σ^2 is the variance in the time distribution assuming that the contribution from the fast neutron pulse can be neglected. This paper deals with an experimental measurement of N and σ^2 as a function of moderator thickness at energies in the region of 100 eV and 1 keV.

The experiment basically consisted of measuring the shape and area of neutron resonances by observing the capture γ -rays from a sample illuminated with a neutron beam leaving various thicknesses of polyethylene moderator. The experimental conditions were chosen so that the widths of the observed resonances were dominated by the moderator effects. A tantalum sample 0.004 ins thick was used to study the 100 eV region and an iron sample 0.040 ins thick for the 1 keV region. A Moxon-Rae detector on a 3.99 meter flight path was used to measure the γ -ray yield as a function of neutron energy.

The results show that the neutron intensity, irrespective of time distribution, is a maximum for a thickness of 1.1 ins polyethylene at ~ 100 eV and 1.0 ins at ~ 1 keV. The full width at half maximum of the time distribution of the neutrons leaving the moderator (i.e. a quantity proportional to σ) increases with increasing moderator thickness and is 112 nsec at ~ 100 eV and 52 nsec in the 1 keV region for a thickness of 1 inch. The figure of merit has been calculated for the two energies and it is found that in each case it is a maximum for ~ 0.75 ins polyethylene. These results are in reasonable agreement with the calculations of Michaudon.

INTRODUCTION

The source of neutrons for a time-of-flight spectrometer in the intermediate energy range (up to ~ 100 keV) is often provided by an accelerator injecting short bursts of charged particles into a heavy element target. Increased neutron intensity can be obtained by surrounding the target with a moderating material, possibly at the expense of an increase in the width of the neutron burst leaving the moderator. The design of a spectrometer is chiefly aimed at obtaining as intense a neutron burst as possible in the shortest possible time. The ability of accelerators to produce shorter and shorter pulses means that the distorting effect of the moderator becomes more and more dominant and it is clear that a knowledge of the effects of the moderator is very desirable if the optimum design is to be achieved. Also if the spectrometer is used to study slow neutron resonances and these are analysed by the "shape" method, then it is very important to know the time distribution of the neutron pulse at the energies corresponding to the resonances.

Groenewold and Groendijk ⁽¹⁾ have calculated the time dependence of the neutrons leaving a semi-infinite moderator, making some reasonable assumptions to simplify the problem. They deduced that for a delta function pulse of neutrons incident on the moderator at time $t = 0$, the distribution of the neutrons leaving with energy E has the form

$$f(x) = \frac{1}{2} x^2 e^{-x} \quad (1)$$

where $x = \frac{t}{T_m}$ and T_m is the mean collision time of a neutron of energy E with a proton in the moderator. For polyethylene, which has a mean free path $\lambda \sim 0.66$ cms. for energies less than a few kilovolts, the full width at half maximum of $f(x)$ is given by $\frac{1.62}{\sqrt{E}}$ μ s (E in eV). As examples, this gives 162 nsec at 100 eV and 51 nsec at 1 keV.

A very comprehensive set of Monte Carlo calculations on the effects of the moderator has been performed by Michaudon ⁽²⁾. In the calculations, the moderator was assumed to have a finite thickness but infinite

lateral dimensions. His results give slightly smaller values for the width, e.g. 135 nsec at 100 eV and 40 nsec at 1 keV for an ingoing neutron energy of 300 keV and a moderator thickness of four mean free path lengths which corresponds to 1.06 ins polyethylene.

Michaudon also deduces that for a given resolution, the quality of a moderator (i.e. a quantity proportional to the count rate of a detector) can be defined by a "figure of merit" F which is given by

$$F = \frac{N}{\sigma^2}$$

where N is the number of neutrons per unit energy interval leaving the moderator and σ^2 is the variance of the time distribution of the neutron pulse. It is assumed that the fast neutron pulse is so short that it makes a negligible contribution to σ^2 . The highest count rate for a given resolution is achieved with the thickness of moderator which will make F as large as possible.

This paper deals with a direct experimental measurement of N and σ^2 at energies in the region of 100 eV and 1 keV for a range of moderator thicknesses.

EXPERIMENT

The experiment basically consisted of measuring the shape and area of neutron resonances by observing the capture γ -rays from a sample illuminated with a neutron beam leaving various thicknesses of moderator. The experimental conditions were chosen so that the widths of the observed resonances were dominated by the moderator effects. This required the widths in time of the natural Doppler broadened resonances to be as small as possible which in turn implied that a short flight path must be used. The remaining contributions to the observed width, namely the fast neutron pulse, the timing channel width and the time resolution of the detectors, were all maintained much smaller than the widths measured.

Fig. 1 shows the experimental arrangement. The beam from the Harwell Electron Linear Accelerator was deflected through 20° on to a 3 ins diameter natural uranium sphere with a 1 mm thick evaporated copper coating. The electron beam entered the target through a re-entrant hole which terminated 2 - 3 radiation lengths from the centre of the sphere. The neutron spectrum produced is reasonably isotropic and has a broad maximum at ~ 300 keV⁽³⁾.

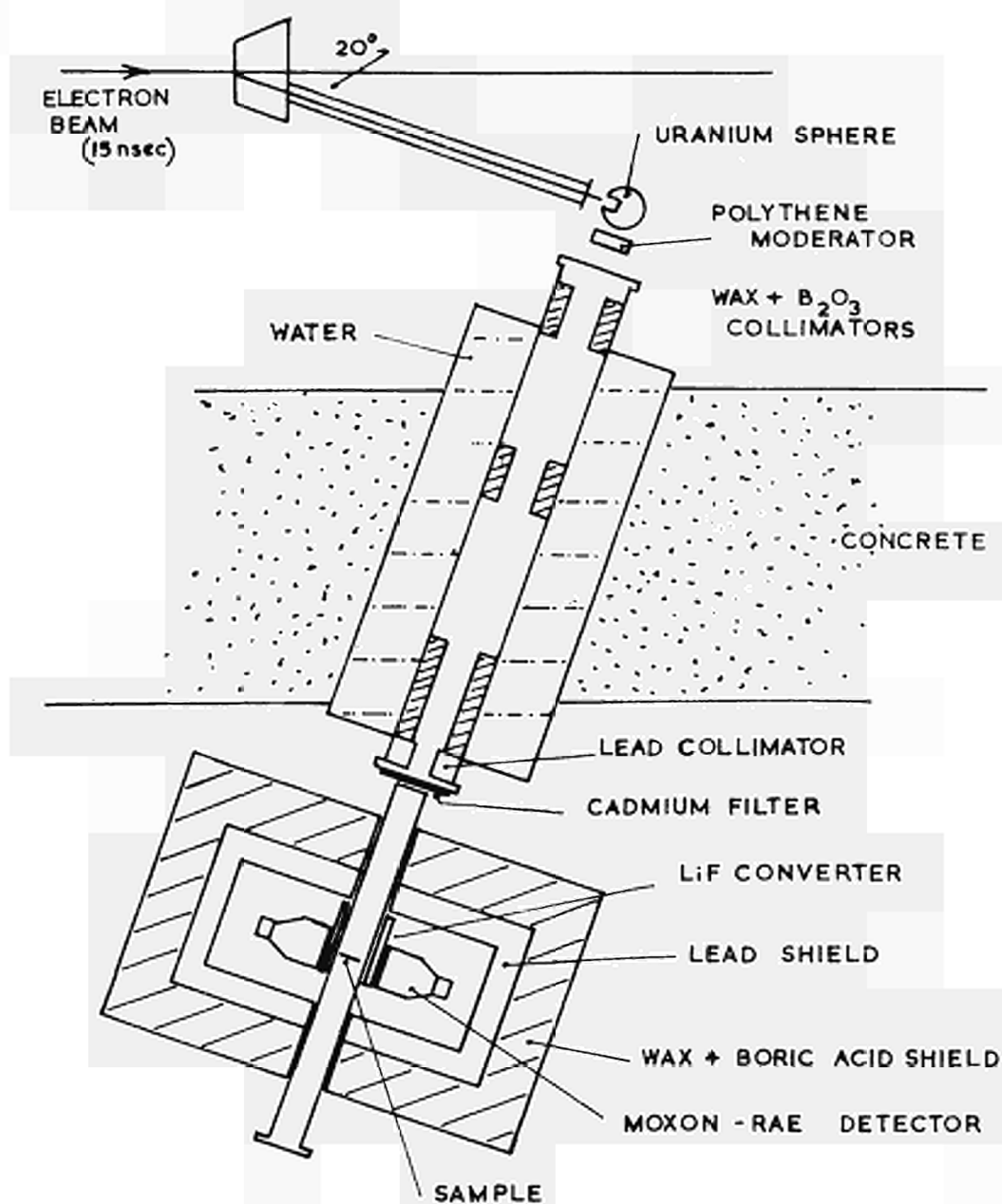


FIG. 1 EXPERIMENTAL ARRANGEMENT

The full width at half maximum of the electron beam was 15 nsec. Measurements by Southan and King⁽⁴⁾ indicate that the broadening of the neutron pulse by multiple scattering in the sphere is negligible. The fast neutron output was monitored by measuring the charge collected by the uranium sphere with a current integrator.

Polyethylene was chosen for the moderator material because of its high hydrogen atom density (7.8×10^{22} H atoms/cm³) and ease of handling. Since no significant resonance peaks were observed with 0.25 inch of polyethylene, seven thicknesses were used, varying from 0.5 in to 2.0 ins in steps of 0.25 in. Each had the form of a 3 ins diameter cylinder with the axis pointing along the flight tube. The moderator face nearest the target was always maintained at a constant distance of 0.125 in from the surface of the sphere and normal to the axis of the flight tube.

Collimators of paraffin wax and boric acid were placed in the evacuated flight tube to produce a 3 ins diameter beam at the sample. The flight path length was 3.99 metres. Where the flight tube passed through the 6 ft thick shielding wall, it was surrounded by 8 ins of water. A sheet of cadmium was placed at the end of the flight tube nearer the detector to remove very low energy neutrons.

The sample was contained at the centre of an evacuated holder 3.5 ft long which passed through an annulus of lithium fluoride 1 inch thick. The lithium fluoride acted as the converter in the Moxon-Rae detector system⁽⁵⁾ used to measure the capture yield. Two counters were used, each consisting of a 5 ins diameter, 0.050 in thick disc of plastic scintillator coupled to a 58 AVP photomultiplier. The detector system was shielded by 4 ins of lead and 6 ins of paraffin wax and boric acid.

Fig. 2 shows the electronic analysing and recording system. The outputs from the counters were added and fed to a fast zero-crossing discriminator. The gain of the system was frequently checked using a ⁶⁰Co γ -ray source which could be placed in a standard, reproducible position. The time expander multiplied the true time-of-flight by ~ 250 so that by feeding the analyser with a 1 Mc/S pulse train synchronised to the firing of the accelerator, channel widths of 4.17 nsec were achieved.

Two samples were used in the experiment. The first was a thin disc of tantalum 0.004 in thick and this allowed measurements to be made simultaneously on four resonances at 99.32, 105.54, 115.08 and 126.46 eV. The second sample used was a disc of pure iron 0.040 in thick and with this sample, the resonance at 1.167 keV was studied.

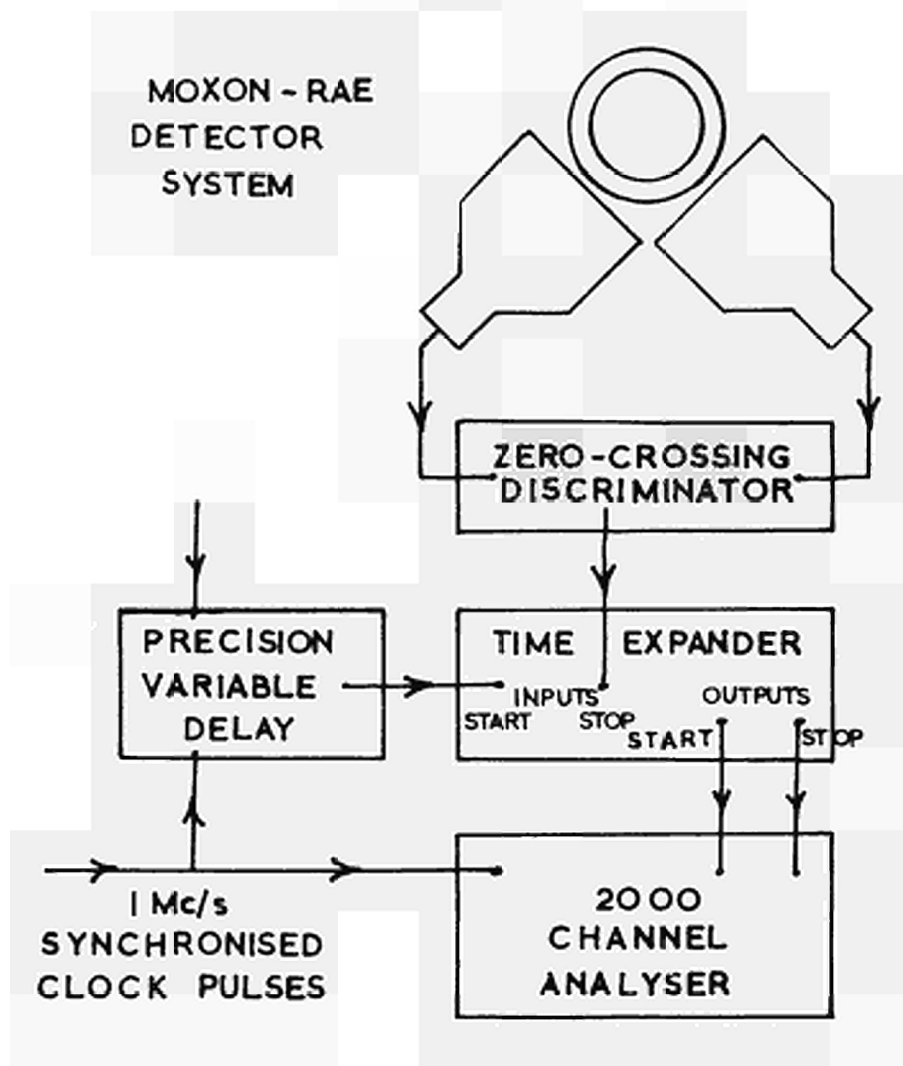


FIG. 2 ELECTRONIC SYSTEM

RESULTS

The results are divided into three sections: (1) neutron intensity, (2) width of the neutron pulse due to the moderator effect and (3) the figure of merit.

(1) Neutron Intensity

After normalisation to unit charge collected on the uranium target, the relative number of neutrons emitted by each moderator thickness is given by the area under the resonance. It should be noted that it is only meaningful to compare results from different thicknesses for the same resonance and not between different resonances. The background level under each resonance was estimated from the flat regions on either side of each resonance.

Fig. 3 shows the results from each of the four tantalum resonances as a function of moderator thickness. It can be seen that the peak in the neutron output in the region of 100 eV occurs with a thickness of polyethylene equal to ~ 1.1 ins. The results of a similar analysis on the 1.167 keV iron resonance are shown in Fig. 4. In this case, the peak occurs at a thickness of 1.0 in.

(2) Time Spread of the Neutron Pulse resulting from the Effects of the Moderator

As previously mentioned, the observed shape of a resonance is determined by a combination of several effects. These include the shape of the fast neutron pulse, the width of the timing channels, the shape of the natural Doppler broadened resonance and the distorting effect of the moderator. The channel width of 4.17 nsec is so small compared to all the other widths that it can be neglected. Although it is not possible to unfold the shape of the neutron burst resulting from moderator effects from the measured shape, we can, to a good approximation, estimate this from the equation

$$(W_{\text{mod}})^2 = (W_{\text{meas}})^2 - (W_{\text{res}})^2 - (W_{\text{fast}})^2$$

where W_{mod} = the pulse width due to moderator effects only

W_{meas} = measured width of resonance

W_{res} = width of Doppler broadened resonance

W_{fast} = width of fast neutron pulse entering moderator.

All widths are measured as full width at half maximum.

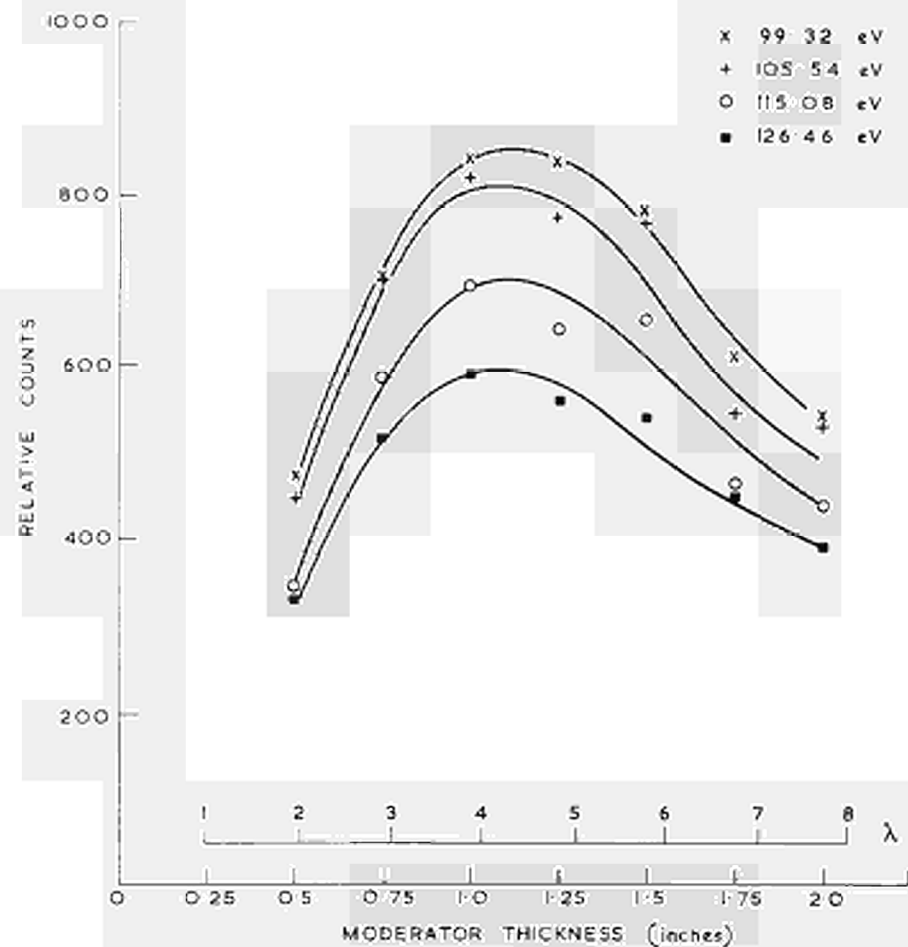


FIG. 3 RELATIVE NUMBER OF NEUTRONS AS A FUNCTION OF MODERATOR THICKNESS FOR THE 100 eV ENERGY REGION

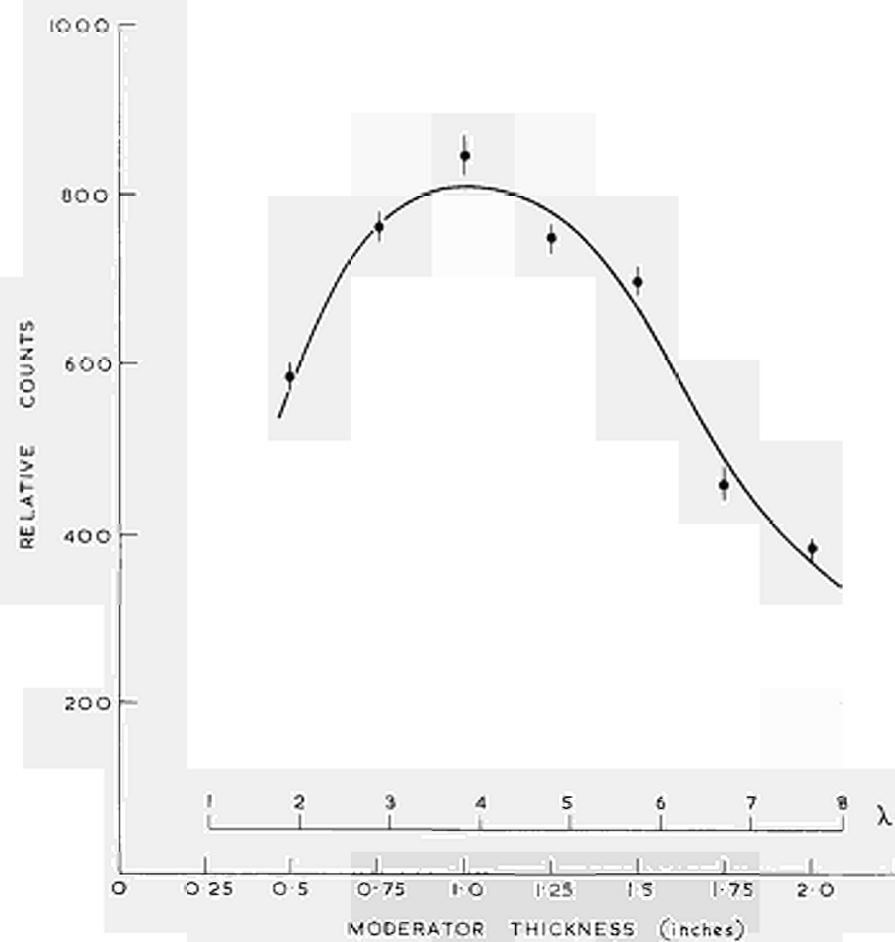


FIG. 4 RELATIVE NUMBER OF NEUTRONS AS A FUNCTION OF MODERATOR THICKNESS FOR THE 1.167 KEV ENERGY REGION

The results of this calculation for the 105.54 and 126.46 eV resonances are shown in Table 1 for the various moderator thicknesses. Curves drawn through the experimental points by eye have been used in estimating the measured widths. The Doppler broadened widths of the resonances were calculated using the resonance parameters given in BNL 325 Vol. II C.

The agreement between the two sets is good (the error in W_{mod} is $\sim \pm 8$ nsec in each case) and it can be seen that in this energy region the time spread introduced by the moderator varies considerably with thickness, being least for the thinnest moderator. The value of ~ 115 nsec at a thickness of 1.06 ins is slightly lower than that given by Michaudon.

Table 1 also shows the results of a similar calculation on the 1.167 keV iron resonance data. At this energy the width W_{mod} varies much more slowly with increasing thickness than at 100 eV.

(3) Figure of Merit

The figure of merit $F = \frac{N}{W_{\text{mod}}^2}$, where N is the relative number of

neutrons shown in Figs. 3 and 4, has been calculated for the various moderator thicknesses at an average energy of 116 eV and also at 1.167 keV. In the 116 eV case, the values of N and W_{mod} obtained from analysis of the 105.54 eV and 126.46 eV resonances were averaged. A smooth curve was drawn through the values of W_{mod} (Fig. 5) and the values taken from the curve were used in the calculation of F. This was done to reduce the error in W_{mod} largely resulting from eye fits to data with ≤ 300 counts in the peaks. The values of F for each moderator thickness are shown in Fig. 6. The maximum value of F occurs with a thickness of ~ 0.75 in polyethylene.

A smooth curve was also drawn through the values of W_{mod} (Fig. 7) obtained from the 1.167 keV resonance and the smooth values used to calculate F. Fig. 8 shows the results and again the peak occurs for a thickness of ~ 0.75 in. Again it must be remembered that no comparison can be made between the values of F shown in Fig. 6 and those in Fig. 8. The thickness of moderator required to make F a maximum at each energy is in reasonable agreement with the calculation of Michaudon.

TABLE I

$$\text{Values of } W_{\text{mod}} = \sqrt{(W_{\text{meas}})^2 - (W_{\text{res}})^2 - (W_{\text{fast}})^2}$$

for various energies and moderator thicknesses.

$W_{\text{fast}} = 15 \text{ nsec}$ in all cases

Energy (eV)	Moderator Thickness (ins)	W_{meas} (nsec)	W_{res} (nsec)	W_{mod} (nsec)
105.5	0.5	104	59	86
	0.75	113		96
	1.0	131		117
	1.25	142		129
	1.5	154		141
	1.75	154		141
	2.0	163		152
126.5	0.5	92	50	76
	0.75	114		101
	1.0	127		116
	1.25	119		107
	1.5	150		140
	1.75	142		132
	2.0	158		149
1167	0.5	48	10	45
	0.75	51		48
	1.0	50		47
	1.25	60		58
	1.5	60		58
	1.75	61		59
	2.0	60		58

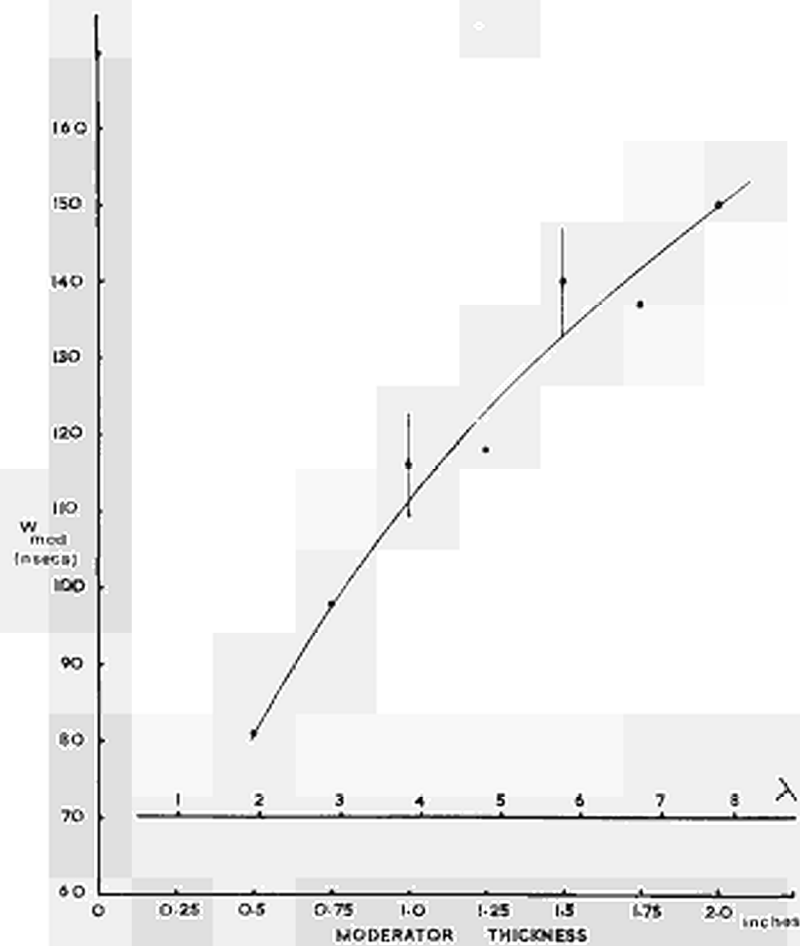


FIG. 5 AVERAGE OF W_{MOD} VALUES FROM 105.5 EV AND 126.46 EV RESONANCES

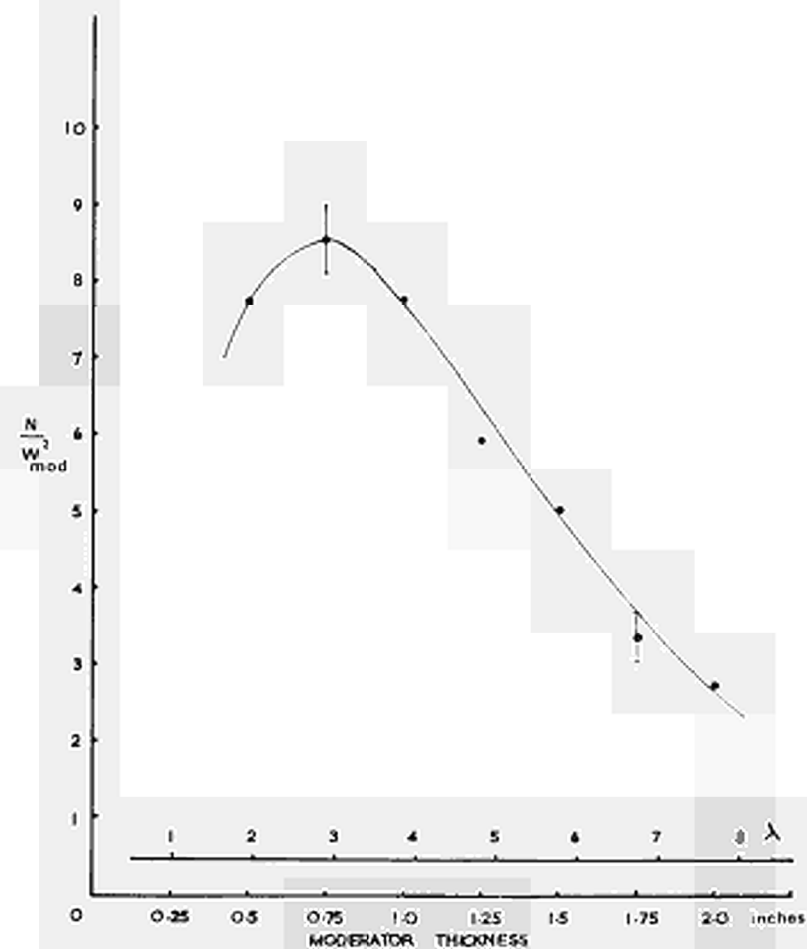


FIG. 6 FIGURE OF MERIT AS A FUNCTION OF MODERATOR THICKNESS USING AVERAGE VALUES OF N AND W_{MOD} FROM 105.5 EV AND 126.46 EV RESONANCES

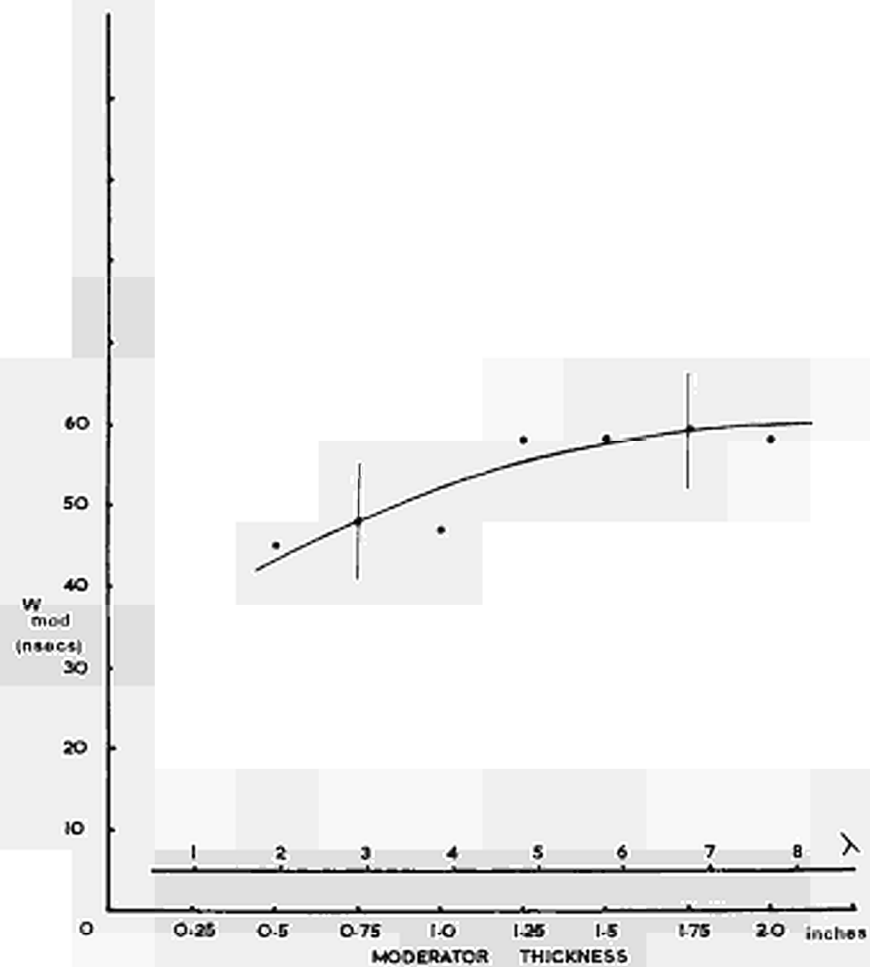


FIG. 7. 1.167 KEV RESONANCE DATA

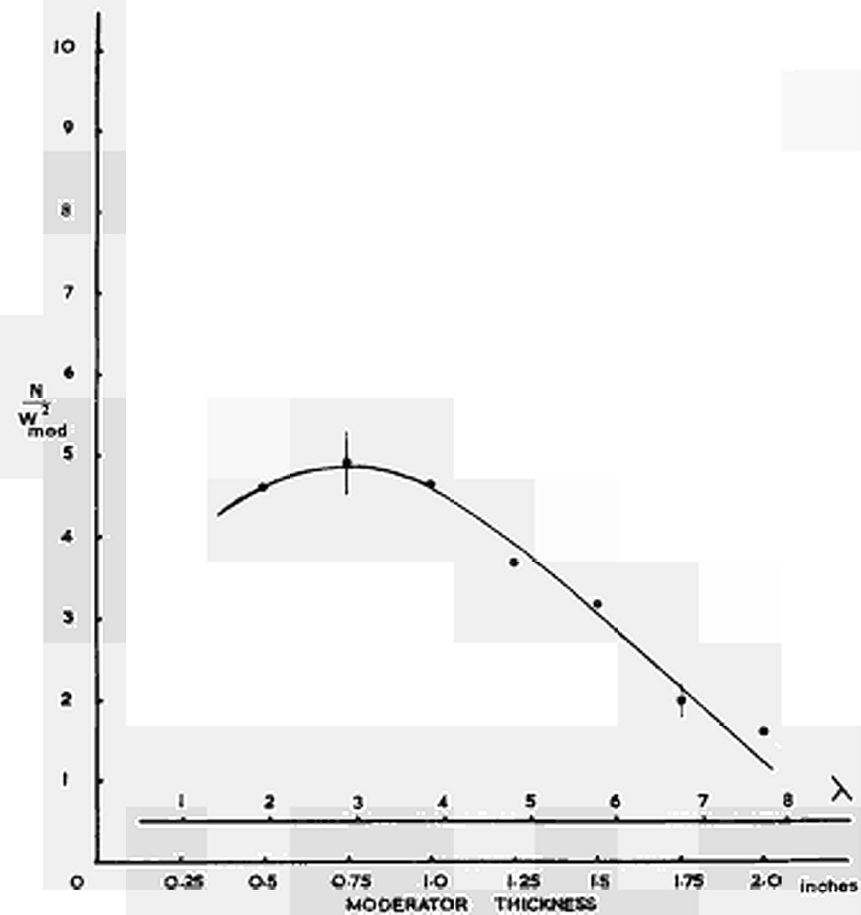


FIG. 8 FIGURES OF MERIT AS A FUNCTION OF MODERATOR THICKNESS USING SMOOTH VALUES FOR W_{MOD} FOR THE 1.167 KEV RESONANCE

CONCLUSIONS

The results of the experiment indicate that the optimum thickness of moderator is about three mean free path lengths (0.78 in polyethylene) at 100 eV and in the region of 1 keV. At this thickness, the corresponding time spreads introduced by the moderator are ~ 100 nsec and 50 nsec respectively. However, in many practical cases, the fast neutron pulse will be comparable to or greater than the moderator effect. Then the shape of the figure of merit curve will become more like that of the relative neutron output shown in Figs. 3 and 4. In such cases, a thicker moderator will be required to give the optimum counting rate.

The shapes of the observed resonances appear to agree reasonably well with that given by equation (1), i.e. the resonances are asymmetric with tails extending to longer delays. This suggests trying to fit equation (1) to the experimental data, using the mean collision time as a variable parameter and folding in the beam, timing channel and Doppler broadened resonances widths. If this is successful, then such an analytic form can be used in the resonance analysis cases requiring knowledge of the distorting effect of the moderator. This fitting procedure is being done at present but as yet no results are available.

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SCHLUSSWORT

O. REIFENSCHWEILER

Philips Research Laboratory, Eindhoven
Netherlands

Da viele von uns an der nächsten "Arbeitstagung über praktische Aspekte der Aktivierungsanalyse mit geladenen Teilchen" am Donnerstag und Freitag nicht mehr teilnehmen, möchte ich einige abschliessende Worte zum Ende der "Arbeitstagung über Beschleunigertargets bestimmt zur Erzeugung von Neutronen" sagen.

Ich meine, dass wir zwei fruchtbare Tage hinter uns haben, in denen wir alle viel gelernt haben. Ausser allgemein interessanten Informationen über Neutronenerzeugung, wie z.B. über Neutronenspektren oder über Optimalisierung von Moderatoren, wurden uns vor allem der neueste Stand der Technik von neutronenproduzierenden Targets und die beim Betrieb solcher Targets gewonnenen Erfahrungen mitgeteilt. Es sei mir erlaubt, etwas zum Stand der Technik dieser Targets, so wie ich ihn selbst nach diesen beiden Tagen sehe, zu sagen.

Wir können bei den neutronenproduzierenden Targets zwei Arten unterscheiden :

1. die in den grösseren Beschleunigern, wie Zyklotrone und Linearbeschleunigern verwendeten Targets, und
2. die in den kleineren kompakten Beschleunigern verwendeten Tritiumtargets.

Bei den Targets der ersten Art handelt es sich zumeist um metallische Targets. Diese bieten wohl keine prinzipiellen Schwierigkeiten, und die sich auf diese Targets beziehenden Vorträge behandelten deshalb vor allem Verbesserungen einer prinzipiell genügenden Wirkungsweise. Einer ganz anderen Situation begegnen wir bei den Tritiumtargets, die in den kleineren Beschleunigern benutzt werden. Die kompakten mit der $^3\text{H} (d,n) ^4\text{He}$ -Reaktiven arbeitenden Neutronengeneratoren sind ja vor allem wegen des relativ kleinen Aufwandes, ihrer Kompaktheit und des relativ geringen Preises von allgemeinerem Interesse.

So warten sehr viele interessante Anwendungen, z.B. die Aktivierungsanalyse in der Industrie, vielleicht auch die Strahlentherapie bösartiger Geschwülste, auf Neutronengeneratoren genügender Ausbeute, Betriebssicherheit und Lebensdauer. Der Realisierung solcher Neutronengeneratoren steht heute eigentlich allein noch das Targetproblem im Wege, da die Erzeugung und Beschleunigung genügend starker Ionenstrahlen als technisch gelöst betrachtet werden kann. Wir können dieses Targetproblem, das wir uns gestellt haben, folgendermassen formulieren:

Es muss ein Target gefunden werden, das eine konstante genügend hohe Neutronenausbeute über eine genügend lange Zeit liefert. Was die Neutronenausbeute anbelangt, denken wir heute an 10^{10} bis 10^{12} Neutronen pro Sekunde, der letzte Wert wird vor allem für eine eventuelle zukünftige Neutronentherapie gefordert. Die Lebensdauer, die wir uns wünschen, ist natürlich so gross wie möglich, auf jeden Fall mindestens einige 10, besser 100, oder gar einige 100 oder 1000 Stunden. Dabei soll das Target noch einfach zu handhaben sein, denn ein einfacher Beschleuniger mit einem schwierig zu handhabenden Target ist eben kein einfacher Neutronengenerator mehr. In diesem Zusammenhang sei vor allem der Gehalt an radioaktivem Tritium nicht zu hoch.

Ich glaube wir sind uns alle darüber einig, dass wir noch weit von der allgemeinen Lösung dieses Targetproblems entfernt sind. Die Technik der konventionellen Tritiumtargets, bei denen das Tritium in exothermen **Wasserstoffabsorbieren** wie Titan, Zirkon oder den seltenen Erden gebunden ist, scheint die Lösung des oben formulierten Targetproblems nicht ohne weiteres möglich zu machen, trotz der Fortschritte auf diesem Gebiet, von denen wir auf dieser Arbeitstagung Kenntnis nehmen durften. Das Tritium wird eben immer noch durch den eindringenden Deuteronenstrahl ziemlich schnell ausgetrieben, und die Neutronenausbeute nimmt im Betrieb viel zu schnell ab. Einen Fortschritt hinsichtlich der Lebensdauer stellen die rotierenden Targets in Aussicht. Ein grosser Nachteil für Routineanwendungen in der Kerntechnik sind jedoch wohl die dabei notwendigen grossen Tritiummengen. So scheint mir, dass zur endgültigen Lösung unseres Targetproblems neue Wege gefunden werden müssen. Ein Versuch hierzu sind gerade die Arbeiten an der hiesigen Universität Lüttich unter Leitung

von Herrn Prof. Govaerts, die die Schaffung eines tritierten Plastik-targets zum Ziel haben. Es kann jedoch bis jetzt noch nicht mit Sicherheit gesagt werden, ob dieser Versuch zu dem erhofften Resultat führen wird.

Eine andere Lösung des Target-Problems bietet im Prinzip die Anwendung von abgeschmolzenen Neutronenröhren, wie wir auf dieser und auch schon auf den früheren Arbeitstagungen erfahren konnten. Bei diesen abgeschmolzenen Neutronenröhren wird durch die Anwendung eines gemischten Deuterium-Tritium-Ionenstrahls mit der Bildung eines Selbsttargets die Lebensdauerbeschränkung durch Austreiben des Tritiums aufgehoben. Konstante Ausbeuten von 10^{10} bis 10^{11} Neutronen pro Sekunde über genügend lange Zeit bieten mit diesen Röhren keine prinzipiellen Schwierigkeiten, und Röhren für 10^{12} Neutronen pro Sekunde wurden uns schon in Aussicht gestellt. Trotz dieses grossen Vorsprungs hinsichtlich der Lösung des Target-Problems und noch anderer Vorteile der abgeschmolzenen Neutronenröhren bin ich der Ansicht, dass eine allgemein anwendbare Lösung des oben formulierten Targetproblems von grosser Bedeutung bleibt. Dies scheint mir vor allem deshalb der Fall zu sein, da der Bau eines an der Pumpe arbeitenden kleinen Beschleunigers sehr viel einfacher ist, als die Herstellung von abgeschmolzenen Neutronenröhren hoher Ausbeute mit guter Betriebssicherheit und langer Lebensdauer. Ausserdem ist ein solcher Beschleuniger in mancher Hinsicht flexibler.

Wir wollen hoffen, dass bis zur nächsten Arbeitstagung weitere wesentliche Fortschritte auf den in den letzten beiden Tagen angedeuteten Wegen oder vielleicht gar auf neuen Wegen gebucht werden können. Es scheint mir im Interesse von uns allen zu sein, dass eine solche Arbeitstagung in zwei bis drei Jahren wieder stattfinden wird.

Falls jemand meine Ausführungen ergänzen möchte oder vielleicht anderer Ansicht ist, möchte ich um Wortmeldung bitten.

Da sich niemals gemeldet hat, darf ich annehmen, dass Sie alle mit dem, was ich gesagt habe, einverstanden sind und vor allem auch, dass Sie in zwei bis drei Jahren wieder eine Arbeitstagung über neutronenproduzierende Targets wünschen.

Ich glaube in Ihrer aller Namen zu sprechen, wenn ich abschliessend unseren Dank all'denen ausspreche, die viel Arbeit für das Zustandekommen

dieser Arbeitstagung geleistet haben. Besonderer Dank gebührt vor allem den Herren, die diese Arbeitstagung vorbereitet haben, den Herren Dr. Ebert und S. Godar von der Kommission der Europäischen Gemeinschaften (EURATOM) und den Herrn Prof. Winand und Prof. Govaerts von der hiesigen Universität und Mitarbeitern. Weiter möchte ich in unser aller Namen den Herren Vortragenden danken, sowie allen Damen und Herren, die in der Diskussion Beiträge geliefert haben.

Hiermit erkläre ich die 4. und letzte Sitzung der "Arbeitstagung über Beschleunigertargets bestimmt zur Erzeugung von Neutronen" für geschlossen.

TEILNEHMERLISTE
LISTE DES PARTICIPANTS
LIST OF PARTICIPANTS

H. ADLER-BARMET	Emile Haefely & Co. Ltd. <u>4000 BASEL - 28</u> Suisse
Ph. ALBERT	C.N.R.S. 15, rue G. Urbain <u>94 VITRY</u> France
Ch. ALLARD	EURATOM BCMN <u>GEEL</u> Belgique
D.A. BANCE	Medical Research Council Radiobiological Research Unit. <u>HARWELL</u> Didcot Berksh. England
E. BARRATT	Rutherford Laboratory Royal Military College of Science <u>SHRIVENHAM NR. Swindon</u> Wiltshire England
R. BERGMAN	D.G. Gustaf Werners Institut <u>UPPSALA</u> Suède
F. BERTRAND	C.E.A. 29-33, rue de la Fédération <u>PARIS 15°</u> France
F. BODART	Université de Louvain Centre de Physique Nucléaire <u>HEVERLEE (Louvain)</u> Belgique
K.H. BOCKHOFF	EURATOM BCMN <u>GEEL</u> Belgique
M. BORMANN	Physikalisches Staatsinstitut I. Inst. für Experimentalphysik <u>2 HAMBURG 36</u> Jungiusstrasse 9 Allemagne
G. BREYNAT	Centre d'Etudes Nucléaires de Grenoble - Section des Accél. B.P. N° 269 <u>38 GRENOBLE</u> France

II

J. BROERSE	Radiobiological Institute TNO 151 Lange Kleiweg <u>RIJSWIJK (ZH)</u>	Hollande
F. BRUNINGHAUS	Brown Boveri & Cie.A.G. <u>68 MANNHEIM - KAFERTAL</u> Postfach 351	Allemagne
E. BRUNINX	Radiochemistry Group Philips Research Laboratories <u>EINDHOVEN</u>	Hollande
L.M. CASPERS	Technological University Physics Dept. Reactor Instituut Berlageweg 15 <u>DELFT</u>	Hollande
H. CEULEMANS	EURATOM <u>MOL</u>	Belgique
F. CORDERO	Junta de Energia Nuclear Avda. Complutense 22 <u>MADRID - 3</u>	Espagne
D. COSSUTA	Multivolt Ltd. 26, Loppets Road <u>CRAWLEY</u>	England
J.E. CROMBEEN	N.V. Philips Research Laboratories <u>EINDHOVEN</u>	Hollande
M. CUYPERS	Laboratoire D'Utilisation Des Radioéléments Université de Liège 9, Place du XX Août <u>LIEGE</u>	Belgique
V. D'ANGELO	C.A.M.E.N. <u>PISA</u>	Italy
S. DEBARROS	Centro Brasileiro de Pq Fisicas Av. Wenceslan Bras 71 <u>Rio de Janeiro</u>	Brésil
H. de CARVALHO	Centro Brasileiro de Pq Fisicas Av. Wenceslau Bras 71 <u>RIO DE JANEIRO</u>	Brésil

G. DECONINCK	Centre de Physique Nucléaire Université de Louvain Parc d'Arenberg <u>HEVERLE - LOUVAIN</u>	Belgique
J. DETAINT	C.E.A. Section des Accélérateurs C.E.N.G. B.P. 269 <u>GRENOBLE</u>	France
H. DILLEMANN	S.A.M.E.S. 21, rue Jean Macé <u>38 GRENOBLE</u>	France
E. DONNEAUX	Physique Nucléaire Laboratoire Van de Graaff Université de Liège 1A Quai Roosevelt <u>LIEGE</u>	Belgique
P. DUMONT	Physique Nucléaire Laboratoire Van de Graaff Université de Liège 1A Quai Roosevelt <u>LIEGE</u>	Belgique
L. DUPAIN	Physique et Chimie Nucléaires Université de Liège <u>LIEGE</u>	Belgique
H.G. EBERT	EURATOM 51, rue Belliard <u>Bruxelles</u>	Belgique
Ch. EGGERMANN	C.E.N. Saclay B.P. n° 2 <u>GIF-sur-YVETTE</u>	France
Ch. ENGELMANN	C.E.A. France C.E.N. Saclay B.P. N° 2 <u>91 GIF-SUR-YVETTE</u>	France
H. FABIAN	Nukem GmbH <u>6451 WOLFGANG b/HANAU</u> Postfach 869	Allemagne
E. FORT	C.E.N. Cadarache <u>St. PAUL-lez-DURANCE</u> B.P. n° 1	France

IV

J. FUGER	Université de Liège Chimie Nucléaire 2, rue A. Stevart <u>LIEGE</u>	Belgique
E.R. GAERTTNER	Linear Accelerator Laboratory Rensselaer Polytechnic Institute <u>TROY N.Y.</u>	U.S.A.
G. GERMAI	Laboratoire D'Utilisation Des Radioéléments Université de Liège 9, Place du XX Août <u>LIEGE</u>	Belgique
F. GIRARDI	C.C.R. EURATOM <u>ISPRA</u>	Italie
Z. GOFFER	Israel Atomic Energy Commission Soreq Nuclear Research Centre <u>YAVNE</u>	Israel
J. GOVAERTS	Université de Liège Laboratoire D'Utilisation Des Radioéléments 9, Place du XX Août <u>LIEGE</u>	Belgique
M. GUILLAUME	Université de Liège Institut de Physique et Chimie Nucléaires 9, Place du XX Août <u>LIEGE</u>	Belgique
M. HUSAIN	Centre de Physique Nucléaire Heverlé-Louvain Atomic Energy Center <u>Ramna - Dacca - 2</u>	Pakistan (East)
C.G. INVERNIZZI	United States Radium Corp. (EUROPA) 36, avenue Krieg <u>1208 GENEVE</u>	Suisse
Ph. JESSEN	Kaman Nuclear <u>COLORADO SPRINGS</u>	U.S.A.
A.H. JIGGINS	Borough Polytechnic Dept. of Physics <u>LONDON S.E.I</u>	England

V

P. KRAMER	Philips Research Labs. <u>EINDHOVEN</u>	Hollande
P. KUIN	Philips Research Labs. <u>EINDHOVEN</u>	Hollande
J. KUTZER	NUKEM <u>645 Hanau</u> Postfach 869	Allemagne
C. LAPERCHE	Laboratoire d'Utilisation Des Radioéléments Université de Liège 9, Place du XX Août <u>LIEGE</u>	Belgique
P.L. LATOUR	High Voltage Engineering (EUROPA)N.V. Amsterdamseweg 61 <u>AMERSFOORT</u>	Hollande
M. LAUGIER	Lab. Central de l'Armement <u>94 ARCUEIL</u>	France
J. LAVERLOCHERE	C.E.N.G. B.P. 269 <u>38 GRENOBLE</u>	France
G. LUCAS	C.E.A. Centre de Limeil B.P. 27 <u>94 VILLENEUVE-SAINT-GEORGES</u>	France
D.A. LUNDBERG	Elliott Electronic Tubes Ltd. Elstree Way <u>Borehamwood Herts.</u>	Angleterre
G. LUTHARDT	Institut für Anorganische Chemie und Kernchemie Universität Mainz <u>MAINZ</u>	Allemagne
P. MALMBORG	The Gustaf Werner Institute Univ. of Uppsala <u>UPPSALA</u>	Suède
F. MANERO	Junta de Energia Nuclear Ciudad Universitaria <u>MADRID - 3</u>	Espagne
F. MANIN	C.E.N. Saclay B.P. 2 <u>GIF-sur-YVETTE</u>	France

VI

S. MARKOWITZ	University of California & Lawrence Radiation Laboratory <u>BERKELEY Calif. 94720</u> U.S.A.
K.W. MARLOW	Instituut voor Kernfysisch Onderzoek Ooster Ringdijk 18 <u>Amsterdam-0</u> Hollande
S. MAY	C.E.N. Saclay B.P. n° 2 <u>91 GIF-sur-YVETTE</u> France
J.P. MEULDERS	Centre de Physique Nucléaire Av. Cardinal Mercier <u>HEVERLE-LOUVAIN</u> Belgique
J.P. MEYER	Centre de Recherches Nucléaires Dept. de Chimie Nucléaire <u>67 STRASBOURG 3</u> France
P. MEYERS	Instituut voor Kernfysisch Onderzoek Oosterringdijk 18 <u>AMSTERDAM</u> Hollande
G. MODIANO	U.S. Radium Corp. <u>GENEVE</u> Suisse
K. MORGANSTERN	Radiation Dynamics Inc. <u>WESTBURY L.I. N.Y.</u> U.S.A.
J. MORIN	C.E.A. 29 - 33 rue de la Fédération <u>PARIS 15°</u> France
M. MOUTHON	SODERN 23, rue du Retrait <u>PARIS 20°</u> France
H. MUNZER	Sektion Physik der Univ. München <u>D-8 MUNCHEN</u> Amalienstr. 54 Allemagne
R. NANQUETTE	C.S.F.-Div. Accélérateurs B.P. n° 10 <u>91 ORSAY</u> France
P.W. NICHOLSON	Physics Dept. Middlesex Hospital Medical School <u>LONDON W.1</u> Angleterre

VII

G. PAIC	Institute Ruder Boskovic P.P. 171 <u>ZAGREB</u>	Yougoslavie
B.H. PATRICK	A.E.R.E. Nuclear Physics Div. <u>HARWELL</u> Didcot	Angleterre
A. PEETERMANS	Université de Liège <u>LIEGE</u>	Belgique
M. PEISACH	Southern Universities Nucl.Inst. P.O. Box 17 <u>FAURE C.P.</u>	South Africa
J.M. PETERS	Laboratoire D'Utilisation Des Radioéléments Université de Liège 9, Place du XX Août <u>LIEGE</u>	Belgique
T.B. PIERCE	Analytical Sciences Division Atomic Energy Research Establ. <u>HARWELL</u>	Angleterre
B. PIRES	Junta de Energia Nuclear Laboratoire de Fisica e Engenharia Nucleares <u>SACAVEM</u>	Portugal
A. PROCUREUR	M.B.L.E. Equipements Philips 80, Rue de deux Gares <u>BRUXELLES</u>	Belgique
L. QUAGLIA	Physique Nucléaire Laboratoire Van de Graaff Université de Liège 1A Quai Roosevelt <u>LIEGE</u>	Belgique
E.R. RAE	A.E.R.E. <u>HARWELL</u> Berksh.	Angleterre
O. REIFENSCHWEILER	Philips Research Laboratory <u>EINDHOVEN</u>	Hollande

VIII

P. REIMERS	Bundesanstalt für Materialprüfung <u>1 BERLIN 45</u> Unter den Eichen 87	Allemagne
J. RETHMEIER	Vrije Universiteit de Boelelaan 1081 <u>AMSTERDAM</u>	Hollande
E. RICCI	Oak Ridge Nat. Laboratory P.O. Box X <u>OAK RIDGE</u> Tenn.31830	U.S.A.
G. ROBAYE	Physique Nucléaire Laboratoire Van de Graaff Université de Liège 1A Quai Roosevelt <u>LIEGE</u>	Belgique
M. ROCHE	C.E.A. 29-33, rue de la Fédération <u>PARIS 15°</u>	France
W. SCHAARSCHMIDT	<u>43 Essen</u> Dreilindenstr. 95	Allemagne
M. SCHMIDT HONOW	Universität Köln Institut für Kernchemie <u>KÖLN-Rhein</u>	Allemagne
F. SCHULZ	Gesellsch. für Kernforschung mbH <u>KARLSRUHE</u> Postfach 947	Allemagne
E. SCHUSTER	Siemens A.G. Forschungslaboratorium <u>852 ERLANGEN</u> 2, G.- Scharowskystr.	Allemagne
M.C. SCOTT	Physics Department The University P.O.Box 363 <u>BIRMINGHAM 15</u>	Angleterre
R. SMEERS	Physique Nucléaire Laboratoire Van de Graaff Université de Liège 1A Quai Roosevelt <u>LIEGE</u>	Belgique
D.L.E. SMITH	A.W.R.E. <u>ALDERMASTON</u>	Angleterre

IX

G. SYLIN	Université Libre de Bruxelles 50, Av. F. Roosevelt <u>BRUXELLES 6</u>	Belgique
M. TALAGRAND	Laboratoire Central de l'Armement <u>94 ARCUEIL</u>	France
Ch. THOMA	Hüttenwerk Oberhausen A.G. Forschungsinstitut <u>42 OBERHAUSEN</u>	Allemagne
R. TIMMER	Philips Gloeilampenfabriek <u>EINDHOVEN</u>	Hollande
A. TRAPANI	EURATOM <u>PETTEN</u>	Hollande
A. TURRIAN	Reaktor Instituut Berlageweg 15 <u>DELFT</u>	Hollande
J. VANDENBROECK	B.I.R.N. 1026-1048 Ch. de Louvain <u>BRUXELLES</u>	Belgique
A. VAN KRANENBURG	Philips Cyclotron Group N.B. <u>EINDHOVEN</u>	Hollande
E. WAIBEL	Physikalisch-Technische Bundesanstalt <u>33 BRAUNSCHWEIG</u> Bundesallee 100	Allemagne
G. WEBER	Laboratoire D'Utilisation Des Radioéléments Université de Liège 9, Place du XX Août <u>LIEGE</u>	Belgique
F.S. WILLIAMSON	Argonne National Laboratory S. Cass Avenue 9700 <u>ARGONNE</u> Illinois 60439	U.S.A.
L.J.F. WINAND	Université de Liège Physique Nucléaire 1A Quai Roosevelt <u>LIEGE</u>	Belgique

X

J.L.D. WOOD

Services Electronics
Research Laboratory
BALDOCK Herts.

Angleterre

P. WUCHERER

A.E.G.
Frankfurt - Main
Goldsteinstr. 235

Allemagne

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